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DIFFUSION AND BIOLOGICAL OXIDATION AS COMPONENT PROCESSES REGULATING THE DEPOSITION OF TRITIATED HYDROGEN TO SOILS

Report No 90-235-K

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ABSTRACT

Deposition of HT to soils was studied in the laboratory using samples from eleven sites in Ontario. The HT deposition velocity, v_g, was measured together with the depth profile of the HTO deposited in the soil and the soil water content, organic content and texture, in order to examine deposition rates in terms of the diffusion of HT into the soil and biological oxidation within the soil. Among individual soil types, deposition velocities decreased as soil water content increased. Measured deposition velocities ranged from 0.13×10^{-4} to 10.2×10^{-4} m s^{-1}. Values for the HT diffusion coefficient in soil, D_{eff}, the local HT oxidation rate in the soil, k_N, and v_g were all found to increase with the fraction of the soil pore volume containing air. This suggests that the factors controlling the access of HT to biological reaction sites within the soil, such as diffusion of HT into the soil and the presence of a moisture film on soil particles, regulate the deposition of HT to soils. Experimental results also verify a theoretical treatment of deposition in terms of D_{eff} and k_N. It is suggested that this theory might provide a framework for predicting v_g on the basis of readily measured soil properties.

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EXECUTIVE SUMMARY

DIFFUSION AND BIOLOGICAL OXIDATION AS COMPONENT PROCESSES REGULATING THE DEPOSITION OF TRITIATED HYDROGEN TO SOILS

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Elemental tritium (T_2, HT, DT; herein referred to as HT) undergoes rapid biological oxidation to tritiated water (HTO) in surface soils, which is the main sink for atmospheric hydrogen and its isotopes. Deposition of HT to soils via conversion to HTO is therefore a significant pathway for the contamination of plants and water by anthropogenic HT sources. In addition, re-emission of HTO from soils to the atmosphere, even in small amounts, essentially determines the inhalation and immersion dose as uptake of HTO by the human body is about 10^4 times more rapid than uptake of HT. Accurate modelling of the deposition of HT to soils is therefore essential to predict reliably the dose resulting from chronic or acute atmospheric HT releases. Even though many measurements of HT deposition velocities have been made, proven methods of predicting deposition velocity on the basis of a knowledge of soil and environmental conditions have not been available for dose impact assessment.

Our objective was to establish the relative importance of gaseous diffusion of HT into soil and subsequent biological oxidation of HT within the soil as component processes of deposition velocity, and to determine how each process was influenced by environmental factors such as soil water content, particle composition and organic content. The experimental approach therefore represents an attempt to understand the mechanisms underlying the deposition process. Deposition velocities for HT and the resulting profiles of deposited HTO were determined in the laboratory for soil cores taken from eleven sites in Ontario.

Deposition velocities (v_g) measured in our study varied between 0.13 x 10^{-4} to 10.2 x 10^{-4} m s^{-1}, consistent with the ranges of values reported from other laboratory and field studies. Values for the HT diffusion coefficient in soil, D_{eff}, the local HT oxidation rate in the soil, k_N, and v_g were all found to increase with the fraction of the soil pore volume containing air. These results confirm the importance of soil water content in controlling the deposition of HT to soil and suggest that deposition velocity is regulated by processes which control the access of HT to reaction sites in the soil, ie, the diffusion of HT into soil and the diffusion of HT in soil pore air to the enzymatic reaction sites. Increases in soil water content restrict the free pore volume in soils, thereby limiting diffusion, and also result in partial to complete encapsulation of soil particles with water films, thereby restricting access of relatively insoluble HT to reaction sites on the soil particle surfaces.

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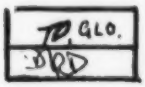
Sufficiently comprehensive measurements were taken to evaluate an earlier theory which describes deposition velocity in terms of the component processes of reaction and diffusion. This theory is currently used in Ontario Hydro's Environmental Tritium Model (ETMOD) to describe HT deposition and the resulting HTO depth profiles in soil. The experimental results were found to be in very good agreement with theory. It was therefore suggested that this theory might provide a framework for predicting v_g on the basis of readily measured soil conditions alone. Further work to determine appropriate methods for predicting effective diffusion coefficients for HT in soil on the basis of soil texture (or bulk density) and soil water content under above-freezing conditions is possible using the results from the present experiment and is recommended in order to improve the ability to predict v_g under natural conditions for the purpose of modelling the dose impact of HT releases.

More generally, the experimental approach used in this study provides a new method of determining diffusion coefficients for gases that react in soil.



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To: Dr. D. Mills
Director of Research

DIFFUSION AND BIOLOGICAL OXIDATION AS COMPONENT PROCESSES REGULATING THE DEPOSITION OF TRITIATED HYDROGEN TO SOILS

1.0 INTRODUCTION

Rapid conversion of elemental tritium (T_2, HT, DT; herein referred to as HT) to tritiated water (HTO) occurs in surface soils and therefore represents a significant pathway for the contamination of plants and water by anthropogenic HT sources (Watts and Murphy 1978, McFarlane et al. 1979). Once formed, HTO in soil is re-emitted to the atmosphere where it can dominate the radiation dose due to inhalation or exposure. HTO behaves similarly to normal water and is taken up by the human body approximately 10^4 times more efficiently than HT (ICRP 1979, Peterman et al. 1985). Accurate knowledge of the rate of conversion of airborne HT to HTO in surface soils is therefore essential to the modelling of environmental tritium behaviour for the purpose of estimating the dose resulting from chronic or acute HT releases to the atmosphere. Quantification of the conversion process under natural field conditions has been a major focus of recent international efforts to improve and verify existing dose models and their input parameters (Burnham et al. 1988, Ogram 1988, Djerassi and Gulden 1988). Previous to this, there have been numerous laboratory and field studies of the deposition of HT to soils.

Microbiological activity at the soil surface is a major factor in the tropospheric H_2 cycle with soil being the main sink for hydrogen and its isotopes (Liebel and Seiler 1976, Schmidt 1974, Ehhalt 1973, Garland and Cox 1980, Murphy et al. 1982, Sweet et al. 1983, Dunstall et al. 1985a, Bunnenberg et al. 1986, Conrad 1988). Oxidation of HT (or H_2) and exchange reactions between HT and water in surface soils result from the activity of ubiquitous soil microorganisms and free soil enzymes (Murphy et al. 1976, 1982, McFarlane et al. 1978, 1979, 1980, Garland and Cox 1980, Conrad 1988, Conrad and Seiler 1981, Conrad et al. 1983, Fallon 1982a, b, Sweet and Murphy 1981, Dunstall et al. 1985a, Popelier et al. 1985). The immediate product of HT chemical conversion within soil is almost exclusively HTO with generally less than 0.1% appearing in the form of organically-bound tritium (Garland and Cox 1980, Sweet and Murphy 1984, Förstel 1986, Bunnenberg et al. 1986, Förstel and Trierweiller 1987).

Compared to soil the sea surface is a negligible sink for H_2 and its isotopes owing largely to their low solubility (Garland and Cox 1980). The uptake and conversion of HT by foliage is also negligible compared to soils (McFarlane et al. 1978, Murphy and Pendergast 1979, Garland and

Cox 1980, Sweet et al. 1983, Sweet and Murphy 1984, Dunstall et al. 1985b, Spencer and Dunstall 1986, Diabaté and Honig 1988, Spencer et al. 1988, Brown et al. 1987, 1988, 1990). Biological conversion of HT may, however, proceed relatively rapidly within at least some types of soil litter with deposition velocities generally in the lower range of those to soil (Spencer and Dunstall 1986, Sweet and Murphy 1984).

Atmospheric oxidation processes result in negligible oxidation of H_2 or HT (Conrad 1988, Jacobs et al. 1979, Garland and Cox 1980). Ogram (1982) estimated that the loss of HT from the lower troposphere due to biochemical oxidation in soils at above freezing temperatures was 10^2 to 10^4 times more rapid than either loss through gas-phase reactions or radioactive decay. Isotopic effects (HT, DT, T_2) on the rates of conversion in soil have not been measured.

Soil moisture content is the most influential variable affecting the deposition velocity, v_g, of HT to soil (Garland and Cox 1980, Sweet and Murphy 1981, Dunstall et al. 1985b, Spencer and Dunstall 1986, Förstel 1986, Bunnenberg et al. 1986, Spencer 1988). Seasonal variation in v_g has been attributed to changes in soil moisture content rather than to variations in temperature (Garland and Cox 1980, Förstel 1986, Förstel and Trierweiler 1987). Higher v_g's tend to occur in soils with low to moderate water content with activity rapidly diminishing as water content increases (Garland and Cox 1980, Dunstall et al. 1985b, Fallon 1982b, Conrad and Seiler 1981, Bunnenberg et al. 1986). The predominant influence of soil water content on v_g implies that the process is diffusion limited, reflecting changes to air-filled porosity, rather than being limited by the number or activity of biological conversion sites (Currie 1961, Brady 1974, Bunnenberg et al. 1986). However, deposition velocities also decrease when soil becomes extremely dry, less than 4% moisture by weight, presumably because biological activity becomes limited (Bunnenberg et al. 1986).

Dunstall et al. (1985a) found a gradual decrease in the capacity for microbial conversion of HT to a soil depth of 160 mm. However, very little HT was found to penetrate intact cores below about 40 mm depth before reacting, hence only a fraction of the HT oxidation capacity in subsurface soil layers was realized. Indeed, most of the HTO resulting from HT oxidation was initially deposited within the surface layers of soil, usually with an approximately exponential decline in HTO concentration with depth to negligible values at depths greater than 100 to 200 mm (Garland and Cox 1980, Dunstall et al. 1985a, Fallon 1982a, Bunnenberg et al. 1986, Förstel 1986, McFarlane et al. 1978, Wiener et al. 1988, Ogram et al. 1988a). In some instances, however, deposition profiles in dry porous surface soils have been found to peak 20 or 30 mm below the soil surface (Brown et al. 1987, 1990; Förstel and Trierweiller 1987; Sweet and Murphy 1981). These profiles may be related to soil composition. Fissures occur in soils that shrink on drying (and swell on wetting) which render them subject to atypical diffusion processes (Currie 1961). Also, if the top layer of soil is very dry, microbial activity may be depressed and diffusion enhanced, allowing HT to rapidly penetrate to lower soil layers. Higher moisture content in the lower soil layers could support higher levels of microbial activity and enhanced HT oxidation.

Organic content has also been reported to influence deposition velocity, possibly by modifying soil structure or by affecting biological activity (Liebel and Seiler 1976, Dunstall et al. 1985b). Organic content and previous land use practices are determinants of both available pore space and pore size distribution in soils and thus can influence diffusion (Brady 1974, Rawls 1983). Soils rich in organic matter also support more abundant populations of heterotrophic microorganisms (Dugnani et al. 1986). Soils with vegetation cover show higher v_g's than bare soils (Liebel and Seiler 1976, Fallon 1982a, Dunstall et al. 1985b). Since deposition to vegetation is negligible,

these results suggest that the presence of root systems could enhance soil porosity (and hence diffusion) and increase microbial activity (LaFavre and Focht 1983).

In summary, experiments in various countries in the last ten years or so have demonstrated that the v_g of HT to soil is usually of order 10^{-4} to 10^{-3} m s^{-1} for soils of typical water content under non-freezing conditions, with values below 10^{-4} m s^{-1} found for very wet soils. Good agreement has been found between chamber measurements in the laboratory and field, and experiments in the open environment (Burnham et al. 1988, Ogram 1988, Ogram et al. 1988b). Measured v_g's decreased with increasing water content (which controls the free pore volume fraction) suggesting that diffusion to reaction sites in soil played an important role in controlling deposition rates. However, methods of predicting v_g values on the basis of a knowledge of soil and environmental conditions have not been available. Ogram (1982) presented a theoretical expression for the soil HT v_g in terms of the effective diffusion coefficient of HT in soil and the local rate of oxidation of HT within the soil. This expression has found use in Ontario Hydro's Environmental Tritium Model, ETMOD (Russell and Ogram 1988), but, previous to the present work, had not been validated.

The objective of the present study was to establish the relative importance of gaseous diffusion of HT into soil and the subsequent biological conversion of HT to HTO as component processes of deposition velocity, and to determine how each process was influenced by environmental factors such as soil moisture content, particle composition and organic content. Deposition profiles for HTO were thus determined for eleven soil types under conditions of varying soil water content. In particular, sufficiently comprehensive measurements were taken to evaluate whether the theoretical expression for HT deposition velocity, discussed above, accurately modelled HT deposition.

2.0 METHODS AND MATERIALS

2.1 Experimental

Intact soil cores were collected from eleven sites within the Ontario counties of Peel, York, Durham and Northumberland. These sites represented a wide range of soil types with respect to particle composition (porosity) and organic content. Following the removal of surface vegetation and stones, soil cores were extracted using polyvinyl chloride cylinders (80-mm ID, 160-mm height). Additional soil samples from each site were analyzed for percent sand, silt and clay, percent organic content and pH by the Soil Testing Laboratory, University of Guelph.

The water content of several soil cores was adjusted following transportation of the chilled samples to the laboratory. Sets of replicate cores corresponding to four different water contents were established. Water was added (X and 2X volumes) to two sets of (2 or 3) replicate cores, one set of replicates was air dried in a fumehood before exposure to HT, and one set was not treated, being left loosely covered. Experiments were initiated the following morning. A single autoclaved soil core was included as a control (ie: no biological activity). The bottoms of the cores were sealed using plastic petri dishes and silicone sealant.

Soil cores were exposed to HT at approximately 20 kBq L^{-1} at 20°C in a 360-L acrylic and PVC chamber over a duration that ranged from 26 to 49 min. The top of the chamber, made of clear plastic, was inserted into a trough that rimmed the bottom portion of the chamber. The two

chamber portions were sealed with 500 mL of distilled water that was added to the trough. Gaseous tritium was injected into the chamber from a pressurized stainless-steel cylinder. Further description of the chamber is given by Dunstall et al. (1985b). During the course of individual experiments the decrease in initial HT concentration varied between 11 and 15%. Tritium-in-air concentrations were measured using an Overhoff tritium monitor that was connected by a closed loop to the chamber. Signal output of the tritium monitor was to a chart recorder. Inventories conducted for each experiment indicated that the loss of tritium was less than 3.5% (Appendix 4) and the amount of tritium in the chamber rim water represented 0.1 to 0.3% of the initial tritium activity. Earlier testing had indicated that the fan located centrally on the top of the chamber created uniform air mixing within the chamber (Dunstall et al. 1985b).

Following HT exposure, the chamber water seal was broken and the chamber air was vented to a fume hood. Each core was immediately sectioned into depth intervals of 0-15, 15-30, 30-60, 60-90 and 90-120 mm. Each of these sections was divided into two equal portions. One portion was immediately analyzed for water content by drying at 105°C to constant weight. To the second portion, 10 mL toluene was added to arrest biological activity and the samples were kept frozen in glass jars for subsequent tritium analysis. Prior to analysis, tritium-free water was added to each sample to give a total water volume, including soil water, of 15 mL, and an additional 40 mL of toluene was added. Soil tritium was extracted by azeotropic distillation (Moghissi et al. 1973) and tritium activity of the samples was determined by liquid scintillation counting.

The deposition velocity, v_g, is defined as the HT deposition flux per unit horizontal area of soil, J_{HT}, divided by the concentration of HT in air, [HT].

$$v_g\ (m\ s^{-1}) = J_{HT}\ (Bq\ m^{-2}\ s^{-1}) / [HT]\ (Bq\ m^{-3}) \qquad (1)$$

vg was determined from the time-integrated form of (1), ie,

$$v_g\ (m\ s^{-1}) = \frac{[HTO]_{dep}\ (Bq)}{[HT]\ (Bq\ m^{-3}) \times A\ (m^2) \times t\ (s)} \qquad (2)$$

where $[HTO]_{dep}$ is the amount of tritium deposited to the soil core, A is the surface area of the exposed core and [HT] is averaged over the exposure period (26 to 49 min).

Soil bulk densities (dry mass per unit volume) were determined by drying the soil samples at 105°C to constant weight. Soil porosity, θ_p, was calculated as a fraction of total soil volume from the bulk density, ρ_b, assuming an average particle density, ρ_p, of 2.65×10^{-3} kg m^{-3} (Brady 1974):

$$\theta_p = 1 - \rho_b / \rho_p \qquad (3)$$

Air-filled pore space (free pore volume fraction, θ_a) was calculated as the soil volume fraction not containing solids or water:

$$\theta_a = \theta_p - \theta_w \qquad (4)$$

where θ_w is the soil water volume fraction. For comparing soils of different composition and water content, the fraction of pore space filled with air, $\theta_{a,p} = \theta_a / \theta_p$, was found to be useful.

2.2 Theoretical

The rate of oxidation of airborne HT in contact with a soil surface is conveniently parameterized by a deposition velocity, as the oxidation process leads to a net flux of HT into the soil. In the open environment, the deposition velocity is defined (see (1) above) in terms of the HT concentration in air at a reference height (usually 1 m) above the surface to which deposition is occurring. The deposition velocity is often expressed as the reciprocal of the resistance to deposition (R) which is the sum of a resistance to turbulent and diffusive transfer through the atmosphere from the reference height to the soil surface, R_a, and a resistance to uptake at the soil surface, R_s:

$$v_g = [R_a + R_s]^{-1} \tag{5}$$

Under natural conditions, resistance to uptake at the soil surface for HT is expected to be much greater than the atmospheric resistance, thus v_g would approximate R_s^{-1} (Ogram 1982). (In general, v_g values much less than 10^{-2} m s^{-1} imply that deposition is controlled by surface uptake). It can be shown (Ogram 1982) that resistance to HT uptake at the soil surface is dependent on the effective diffusion coefficient, D_{eff}, of HT in soil and the local reaction rate coefficient for HT oxidation, k, where k and D_{eff} are defined at depth $z=z'$ as follows:

$$J_z\,(z=z') = - D_{eff} \frac{\partial [HT]_z}{\partial z} \Bigg|_{z=z'} \tag{6}$$

$$k\,(z=z') = - [HT]_z^{-1} \frac{\partial [HT]_z}{\partial t} \Bigg|_{z=z'} \tag{7}$$

where J_z is the flux of HT per **unit area of soil** at soil depth z, and $[HT]_z$ is the concentration of HT in air within the soil pores (defined **per unit volume of air**) at depth z. For uniform soil conditions (ie, D_{eff} and k unvarying with depth), theory (reviewed in Appendix 1) predicts an exponential decline in the HT concentration in soil air with depth:

$$[HT]_z = [HT]_z(z=0)\ \exp\,(-z/z_e) \tag{8}$$

where $[HT]_z(z=0)$ is the concentration of HT in air at the soil surface (equal to the concentration in chamber air, [HT]), and z_e is the exponential scaling depth for the HT depth profile (ie, at z_e the HT concentration is 1/e that at the surface).

The rate of HT diffusion into soil is usually sufficiently rapid for a steady state HT profile to be established in minutes, or less (Appendix 1). Diffusion of the product HTO, which is largely incorporated into soil water, is expected to be much slower as liquid-phase diffusion coefficients are orders of magnitude smaller than gas-phase diffusion coefficients (Garland 1980). In our

experiments, the time scales are such that the HT profile would be expected to be near steady state for most of the approximately 30 min exposure period, while the profile of deposited HTO measured in cores sectioned immediately after exposure would reflect the HT profile (Appendix 1). If the concentration of HT in chamber air is constant, then

$$[HTO]_z = k\,[HT]_z\,\theta_a\,\Delta t \quad (9a)$$
$$= k_N\,[HT]_z\,\Delta t \quad (9b)$$

where:

$[HTO]_z$ is the concentration of HTO per unit volume of **soil** at depth z;
$[HT]_z\,\theta_a$ is the concentration of HT per unit volume of **soil** at depth z;
Δt is the duration of the HT exposure;
θ_a is the free pore volume fraction (air-filled pore space; equation 4);
k is the (first-order) rate coefficient for conversion of HT to HTO in soil; and
$k_N = k\,\theta_a$ is the rate coefficient for formation of HTO (as $[HTO]_z$ is expressed per unit volume of soil and [HT] is defined per unit volume of air-filled pore space).

Immediately after the exposure, the deposited HTO profile would have a form similar to the steady-state HT profile established during the exposure period, ie,

$$[HTO]_z = [HTO]_z(z=0)\,\exp(-z/z_e). \quad (10)$$

By using the boundary condition that the deposition flux at the soil surface should be equal to the diffusive flux of HT into the soil, the following relationships can be derived between the variables v_g, D_{eff}, k_N and z_e (Appendix 1):

$$v_g = k_N\,z_e \quad (11a)$$
$$= (k_N\,D_{eff})^{0.5} \quad (11b)$$
$$= D_{eff} / z_e \quad (11c)$$

The parameters v_g, z_e and k_N were determined directly from the present experiments: v_g from (2); z_e by fitting an exponential profile to the depth profile of the HTO deposit; and k_N by extrapolating the depth profile of deposited HTO back to the surface, z = 0, and using the relationship

$$k_N = \frac{[HTO]_z(z=0)}{[HT]\,\Delta t} \quad (12)$$

where the units of $[HTO]_z$ and [HT] are radioactivity per unit volume of soil and air respectively. The units of k_N are reciprocal time (s^{-1}). In reality, estimates of k_N from the soil HTO profile may slightly underestimate the true k_N since the influence of diffusion through a very thin layer at the soil surface may not have been totally eliminated.

D_{eff} was not measured directly, but may be calculated from the bulk density and soil water content using one of the empirical relationships available from the literature. The rate of gaseous diffusion in a soil is known to be related to air-filled pore space, the tortuosity of the diffusion passages and properties of the gas (Pritchard 1985, Currie and Rose 1985, Nielson et al. 1984, Troeh et al. 1982, Currie 1961). Pore structure, and hence tortuosity are dependent on the geometrical properties of the soil. Air-filled pore space is the difference between total porosity and water-

filled pore space (4). Materials with similar porosity may, however, exhibit different relationships between diffusion and moisture content (Currie 1961).

The following empirical relationship, first proposed by Penman in 1940 for the effective diffusion coefficient in soil (Currie 1960; Jacobs et al. 1979), has found widespread use because of its simplicity:

$$D_{eff} = (D_o\ \theta_a)/\tau^0 \qquad (13)$$

where D_o is the diffusion coefficient for HT in air (see below), θ_a is the fractional air-filled pore volume (vol. air/vol. soil) and τ^0 is tortuosity which has an assumed value of 1.5 (Jacobs et al. 1979). This relationship does not take into account the influence of pore structure on the tortuosity of different soils (Troeh et al. 1982) and, while adequate for most soils of low to moderate water content, tends to overpredict diffusion coefficients at high soil water contents. In moist soils, the micropores are the first to fill with water This leads to restriction of diffusion even though air still exists within the macropores (Brady 1974). Diffusion also tends to be lower in soils with a wide range in grain size, and hence a wide range in pore sizes, because of the increase in tortuosity (Nielson et al. 1984). Tortuosity values are useful as soil structure indices but do not always provide a realistic index for predicting diffusion coefficients for different soils. The usual practice is to measure diffusion as opposed to attempting to determine the tortuosity of complex materials such as soil (Troeh et al. 1982).

Our experiments offered the opportunity to compare values of D_{eff} calculated from (13) to values inferred from v_g and k_N, or v_g and z_e (equations 11b and 11c).

The diffusion coefficient for hydrogen in air, D_o, at STP is 0.634×10^{-4} m^2 s^{-1} (Weast and Astle 1982). The D_o for H_2 at room temperature (20°C) is 0.718×10^{-4} m^2 s^{-1}; as derived from the equation of Jellick and Schnabel (1986):

$$D_o(293K) = D_o(273K)\ (T/273)^{1.75}. \qquad (14)$$

Assuming that the diffusion coefficient is proportional to the inverse square root of molecular weight, D_o for HT at 20°C is 0.51×10^{-4} m^2 s^{-1}.

3.0 RESULTS

3.1 Soil Properties

Physical and chemical composition of the 11 test soils were quite different with sand contents that varied between 11 and 88% and organic contents that varied between 0.5 and 12.8% by weight (Table 1). The textural classification of the soils varied from sand to silty clay. All the soils were slightly alkaline (pH 7 to 8).

The pore space of the soils varied from 44 to 73%. Since soil bulk density affects both porosity and water retention of soils, the variation in measured bulk densities for replicate soil cores was examined. Among replicate cores (8 or 9 per site) the coefficients of variation for bulk density ranged from 3.9 to 11.2% with a mean C.V. for all soils of 6.8% (Table 2). Thus there was only slight variation in bulk density among individual soil types. In the Pickering, Bolton, North

TABLE 1

PHYSICAL AND CHEMICAL CHARACTERISTICS OF SOILS USED IN TRITIUM DEPOSITION STUDIES

SITE	TEXTURE	% ORGANIC CONTENT	%CLAY <2 µm	% SILT 2-53 µm	% SAND 53-2000 µm	pH
Cherrywood (C)	Silty Clay	10.5	20	29	52	7.8
Wesleyville (W)	Loam	5.2	16	33	51	7.8
Darlington (D)	Loam	6.6	26	47	28	7.6
Pickering (P)	Loam	9.8	19	39	42	7.1
Bowmanville (O) (Organic)	Sandy Loam	5.3	11	20	69	7.8
Bowmanville (S)	Sand	0.5	2	9	88	8.0
Bolton (B)	Silty Clay	10.5	41	48	11	7.4
Claremont (L)	Clay Loam	12.8	35	45	21	7.3
Glen Major (G)	Loam	11.1	13	36	51	7.5
Ballantrae (A)	Sand	3.5	5	7	88	7.6
Kinsale (K)	Loam	9.0	14	46	39	7.5

TABLE 2

MEASURED BULK DENSITIES AND COEFFICIENTS OF VARIATION FOR ELEVEN SOILS (0 to 120 mm DEPTH)

Site	Bulk Density x 10^3 (kg m^{-3}) Mean	S.D.	n	Coefficient of Variation (%)	Pore Space, θ_s (Volume Fraction)
Cherrywood (C)	0.719	0.058	9	8.1	0.73
Wesleyville (W)	1.36	0.064	9	4.7	0.49
Darlington (D)	1.09	0.088	9	8.1	0.59
Pickering (P)	0.913	0.055	9	6.0	0.66
N. Bowmanville (Organic, O)	1.06	0.119	9	11.2	0.60
N. Bowmanville (Sand, S)	1.49	0.081	9	5.4	0.44
Bolton (B)	0.902	0.056	9	6.2	0.66
Claremont (L)	0.850	0.033	8	3.9	0.68
Glen Major (G)	0.769	0.064	8	8.3	0.71
Ballantrae (A)	1.09	0.043	8	3.9	0.59
Kinsale (K)	0.956	0.082	8	8.6	0.64

Bowmanville (sand) and Ballantrae soils there was a tendency for bulk density to increase with increasing depth while in the remaining soils there were no consistent depth related differences in bulk density within 120 mm of the soil surface. Mean bulk density values for individual sites (0-120 mm average), and corresponding mean porosities, were used for all subsequent calculations.

The influence of soil organic matter on bulk density for several soils is shown in Figure 1. A similar relationship exists for organic matter and percent pore space since the latter is dependent on bulk density (equation 3). Since these variables are interrelated, any one may be used to describe aspects of soil structure relevant to gaseous diffusion. The parameter chosen for subsequent analysis was the fraction of pore space filled with air, $\theta_{a,p}$, as this parameter appeared to scale data better than other variables tested (soil water content, total pore volume fraction, free pore volume fraction, etc).

3.2 Soil HTO Depth Profiles

Typical deposition profiles for tritium were characterized by a near-exponential decline in HTO concentration with soil depth (eg, Figure 2). The HTO concentration at the soil surface was inferred from the y-intercept while a regression fit to the log-linear plot was used to derive the exponential scaling depth of the profile, z_e. Correlation coefficients for ln [HTO] versus depth exceeded -0.92 for all cores except two that had been taken at Wesleyville (2X water). In these two cores there was an abrupt increase in tritium concentration in the deepest section (90 to 120 mm) which indicated that air leakage had occurred through the seal on the bottom of the core. Although one core from Darlington and all cores from the North Bowmanville sandy loam location deviated from the characteristic exponential decline in HTO concentration with soil depth, the correlation coefficients for these samples were reasonable (better than -0.92). In five of these cores maximum HTO concentrations occurred at the 30-60 mm depth while in the other four cores HTO concentrations were similar in the 0-30 mm and 30-60 mm sections.

Averaged over all 88 cores, the amount of tritium deposited in the upper 30 mm of soil represented 74.6% (coefficient of variation, C.V. = 13.7%) of the total amount of tritium deposited, while 93.5% of the total tritium was deposited within 60 mm of the soil surface (C.V. = 5.8%). These values both correspond to an exponential scaling depth, z_e, of about 22 mm (equation 8). With the North Bowmanville sandy loam location excluded, the amounts of tritium deposited within the 0-30 mm and 0-60 mm depths were 76.5% (C.V. = 11.1%) and 94.7% (C.V. = 3.71%), respectively, only slightly different from the values with these samples included.

The depth of HT penetration into the soil is expected to depend on the residence time of HT in soil prior to reaction and thus will decrease with increasing soil reaction rate and increase with HT diffusion coefficient in soil. Figure 3 shows z_e plotted against the fraction of pore space with air, $\theta_{a,p}$. It is striking that nearly all values fell between about 15 and 30 mm (mean 23.9 mm, SD 9.7 mm, geometric mean 22.7 mm, GSD 1.34). The anomalous North Bowmanville sandy loam soil (labelled "O" in the Figure) provided the exception, exhibiting z_e values up to 90 mm. Excluding the North Bowmanville results, there was little trend with $\theta_{a,p}$.

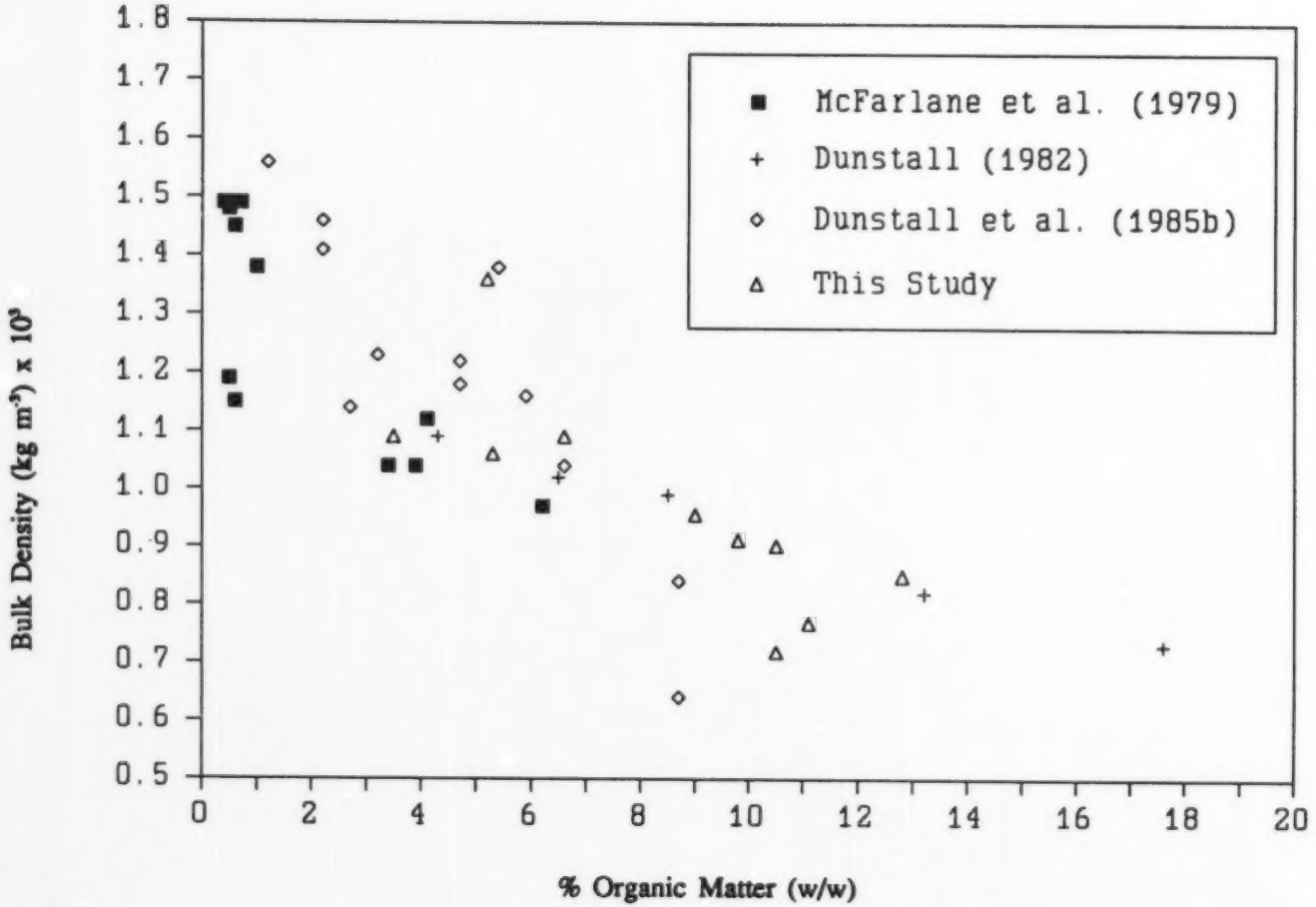

Figure 1. Relationship between bulk density and soil organic content.

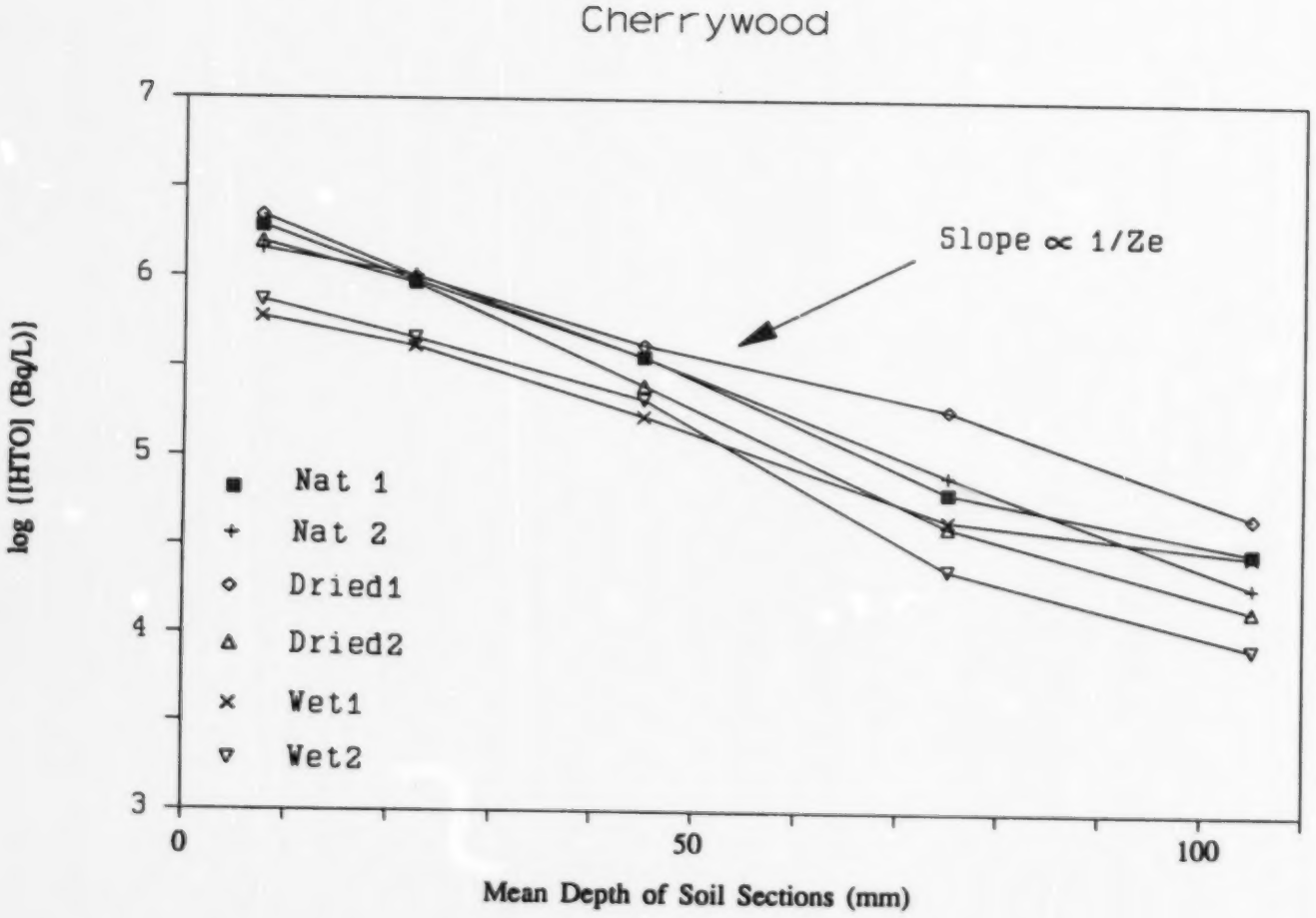

Figure 2. Concentration of HTO in soil water with depth for Cherrrywood cores (Natural, Air Dried and Water Added: duplicates).

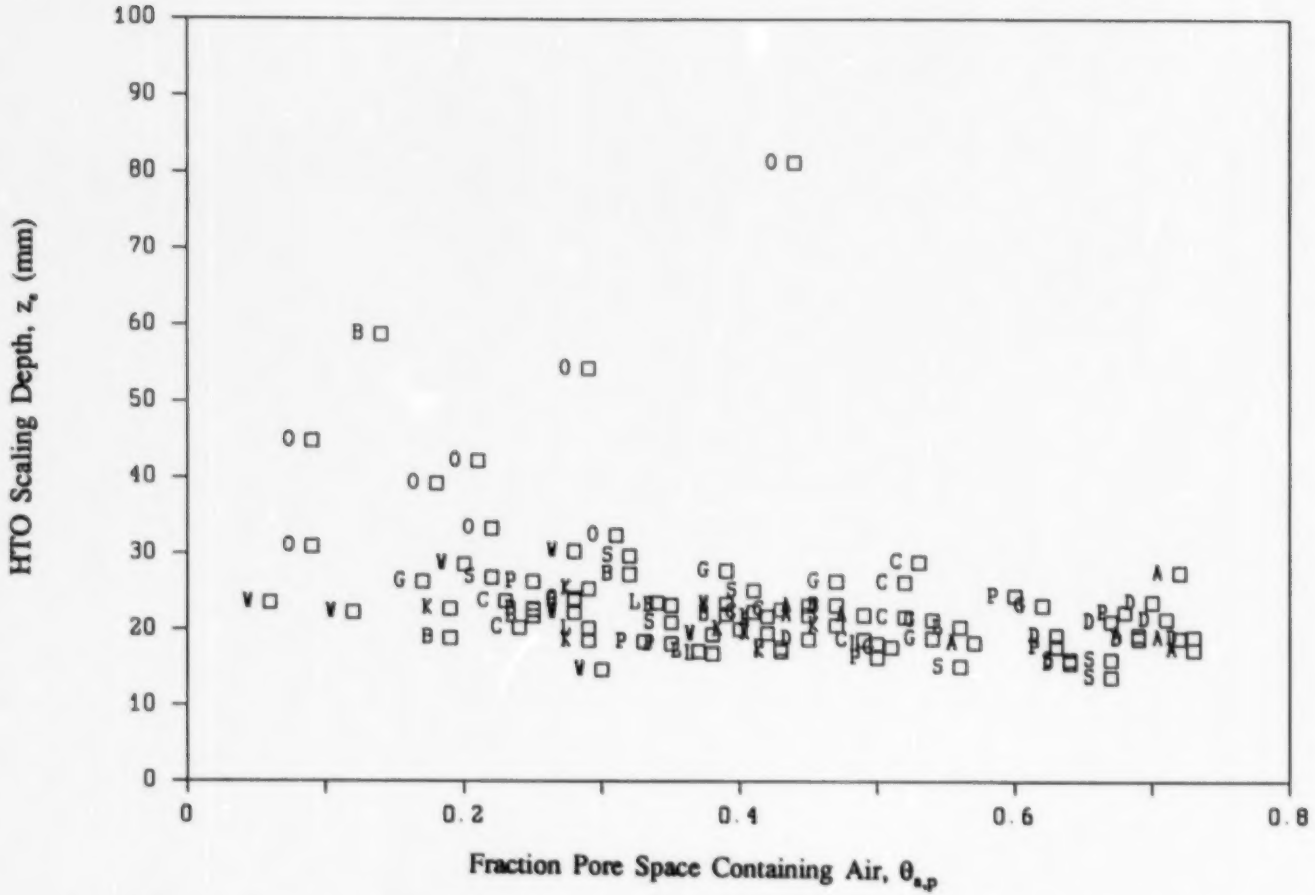

Figure 3. Exponential scaling depth, z_s, for depth profile of HTO deposit versus fraction of soil pore space containing air. Labels refer to soil sampling site (Table 1).

3.3 Soil HT Deposition Velocity

Measured deposition velocities for all soil cores ranged from 0.13 x 10^{-4} to 10.2 x 10^{-4} m s^{-1}. Anomalous v_g's, with maximum values of 1.2 x 10^{-4} m s^{-1}, appeared to have been obtained for the North Bowmanville (organic) sandy loam. This soil did not appear to be exceptional with respect to any of the measured soil parameters and no explanation for the low deposition velocities is apparent. The addition of increasing amounts of water to soil cores resulted in large reductions in v_g. In very moist soils v_g's were comparable to values obtained with sterilized soil samples. Deposition velocity also increased with increasing free pore volume. Figure 4 shows a plot of v_g versus $\theta_{a,p}$. Water content of soils varied from 4% to 84% (of dry weight). The fraction of pore space containing air, $\theta_{a,p}$, ranged from 0.073 to 0.76.

3.4 Comparison of Experiment with Theory

The validity of the theory described earlier (Ogram 1982, Russell and Ogram 1988) was tested by comparing measured deposition velocities with deposition velocities calculated from the independently measured variables k_N and z_e (equation 11a). Deposition velocity was also calculated from D_{eff} and k_N (equation 11b). The effective soil HT diffusion coefficient, D_{eff}, was derived using the correlation with free pore volume fraction given earlier (equation 13), with the free pore volume calculated from the measured bulk density and water content (equations 3,4).

Good agreement, generally within a factor of two, was obtained between experimentally derived and calculated v_g's (Figure 5). The mean and geometric mean (GM) ratio of calculated to measured values, and the standard deviation (SD) and geometric standard deviation (GSD), were also calculated (Table 3). The GM and GSD are probably more appropriate for describing the variation in the ratio of predicted to measured values, as this ratio cannot be negative. The GSD, or a multiple of the GSD, gives directly the fraction of results within a certain factor of the GM. For example, the expression $v_g = k_N z_e$ gives a GM of 1.01 indicating extremely good agreement on average with measured results and a GSD of 1.25 indicating that approximately 95% of calculated values are within a factor of 1.55 (equal to the GSD^2) of the measured value. Detailed results are summarized in Appendix 3.

3.5 Soil HT Oxidation Rate Coefficient

The rate coefficient for biological oxidation at the soil surface, k_N, ranged from 2.6 x 10^{-4} to 6.7 x 10^{-2} s^{-1} (the rate coefficient for conversion of gas phase HT, $k = k_N/\theta_a$, ranged from 2.9 x 10^{-3} to 2.3 x 10^{-1} s^{-1}). By comparison, the variation in v_g was two orders of magnitude and there was a 20-fold variation in calculated diffusion coefficients. Organic content of the soils did not account for variations in k_N between soil types. A plot of log k_N versus the fraction of pore space filled with air, $\theta_{a,p}$, showed reasonably good correlation (Figure 6). For all soil types tested, and for $\theta_{a,p}$ greater than about 0.5, k_N was relatively constant, with values ranging between 0.011 and 0.067 s^{-1} (mean 0.034 s^{-1}, SD 0.013 s^{-1}). Between $\theta_{a,p} = 0.5$ and $\theta_{a,p} = 0.1$, k_N decreased by about two orders of magnitude with decrease in $\theta_{a,p}$ due to increase in water content.

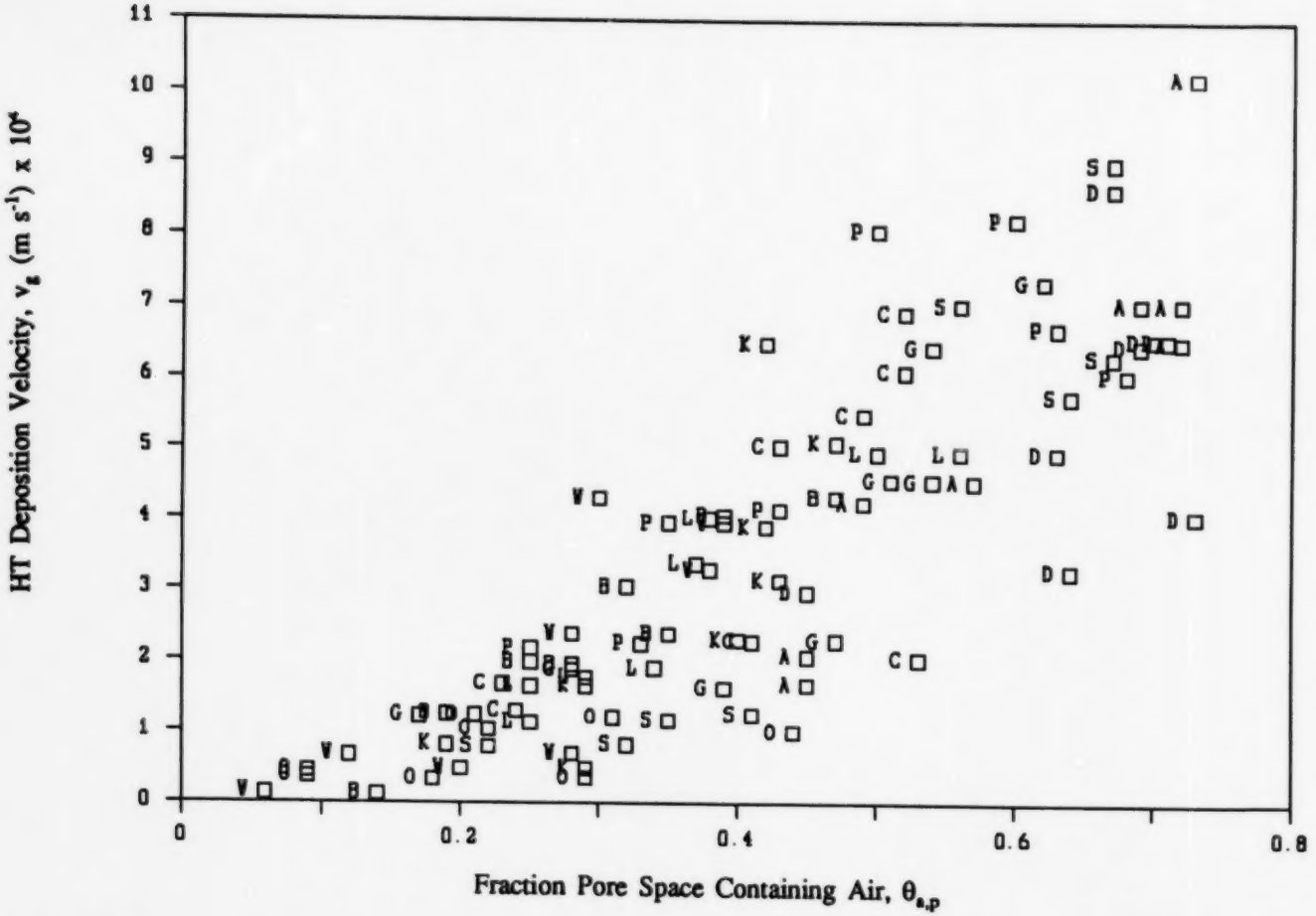

Figure 4. Relation of HT deposition velocity to fraction of pore space containing air for 88 soil cores. Labels refer to soil sampling site (Table 1).

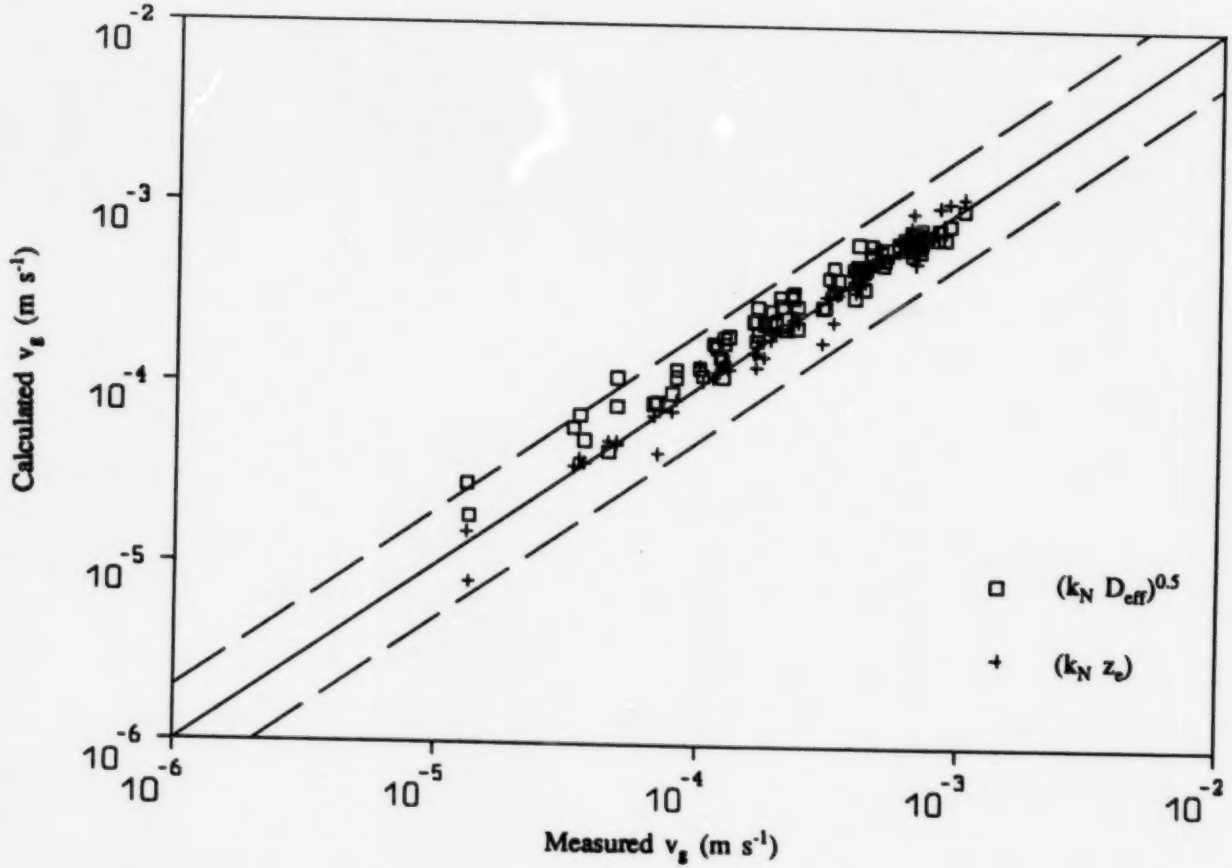

Figure 5. Comparison of experimentally measured deposition velocities of HT to soil to values calculated from k_N, z_e and D_{eff}. The solid line represents a ratio of unity and the dashed lines represent ratios of 2 and 0.5, respectively.

Table 3

Ratio of Calculated to Measured HT Deposition Velocity to Soil (88 data points)

Ratio	Min	Max	Mean	Standard Deviation	Geometric Mean	Geometric Standard Deviation (GSD)
$k_N z_o / v_g$	0.60	1.45	1.02	0.13	1.01	1.24
$(D_{eff} k_N)^{0.5} / v_g$	0.79	2.27	1.20	0.28	1.17	1.15

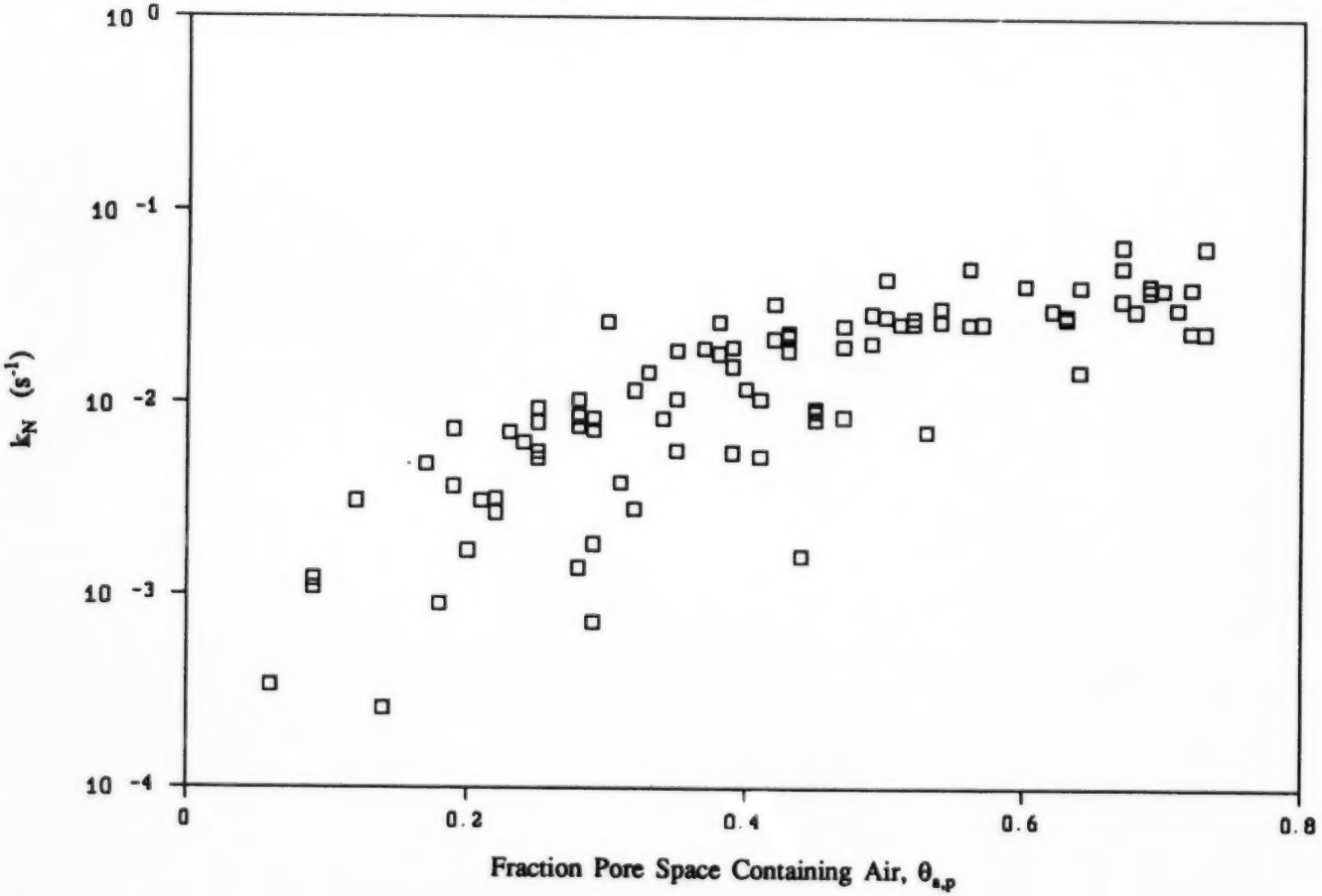

Figure 6. Relationship between rate of biological oxidation of HT in soil, k_N, and fraction of pore space filled with air, $\theta_{a,p}$.

3.6 Soil HT Diffusion Coefficient

The soil HT diffusion coefficient was not measured directly, but can be derived from any two of the directly measured variables v_g, k_N and z_c. Values of D_{eff} were calculated using equations 11b and 11c and compared to each other and to D_{eff} values determined from the empirical relation based on free pore volume with an assumed tortuosity of 1.5 (equation 13).

The D_{eff} values calculated from the experimentally measured parameters v_g and z_c, and v_g and k_N, respectively, were in excellent agreement (Figure 7, Table 4). However, both these methods gave lower values, by about 20 to 30% on average, than the empirical correlation based on soil porosity and tortuosity (Table 4). Figure 7 shows that D_{eff} calculated empirically (13) overpredicted effective diffusion rates by a greater degree at lower values of D_{eff}, ie, at lower free pore volume (which generally corresponded to higher water content).

4.0 DISCUSSION

Measured deposition velocities (v_g) for HT to soil in the present study fell within the range 0.13 x 10^{-4} to 10.2 x 10^{-4} m s^{-1}. These values are consistent with those reported in the literature. Measurements of v_g have been made for homogeneous soil preparations (Fallon 1982a,b, Dunstall et al. 1985a, Spencer and Dunstall 1986), undisturbed soil cores incubated under laboratory conditions (Fallon 1982a, Dunstall et al. 1985a, Förstel 1986), in the field using exposure chambers (Garland and Cox 1980, Sweet and Murphy 1981, Dunstall et al. 1985a, Conrad and Seiler 1981) and in areas exposed to atmospheric HT releases (Sweet and Murphy 1981, Ogram 1988, Wiener et al 1988, Brown et al. 1990).

Most measurements are made using soil samples or areas of limited size. The question arises as to how representative these measurements are of deposition velocities averaged over larger areas. Förstel (1986) found a 35% variation in v_g for an entire field and 14% variation within 1 m^2. Wiener et al. (1988) reported a tenfold variation in v_g measurements within a field and a threefold variation within a plot of thoroughly mixed soil. These variations were attributed to local in-homogeneities in soil conditions and vegetation cover. In the present experiment, little variation was found in bulk density measurements among replicate soil cores. In fact relative consistency in soil texture, bulk density and water content can be expected to exist within a given field (Nielsen et al. 1973). These parameters would therefore appear to be appropriate for the estimation of deposition velocity.

The influence of temperature or other variables that might be expected to affect biological activity would be rate limiting to deposition only if the sites of conversion were fully saturated with HT, and evidence from a number of sources suggests that this is not the case except at extremes of temperature. Soils have been found to exhibit only slight variation in HT deposition rate over a wide range of temperature indicating that the availability of reaction sites was not a primary rate determining factor (Fallon 1982b, Förstel 1986, Garland and Cox 1980, Liebel and Seiler 1976, Dunstall 1982). In general, the rate of HT oxidation has been found to vary by a factor of about two in response to temperature changes over the normal physiological range with maximum oxidation activity between 20°C and 30°C. It has also been shown that full capacity for HT oxidation was not realized in the sub-surface strata of natural soils as the HT reacts before penetrating to depths greater than a few tens of millimetres into the soil (Dunstall et al. 1985a).

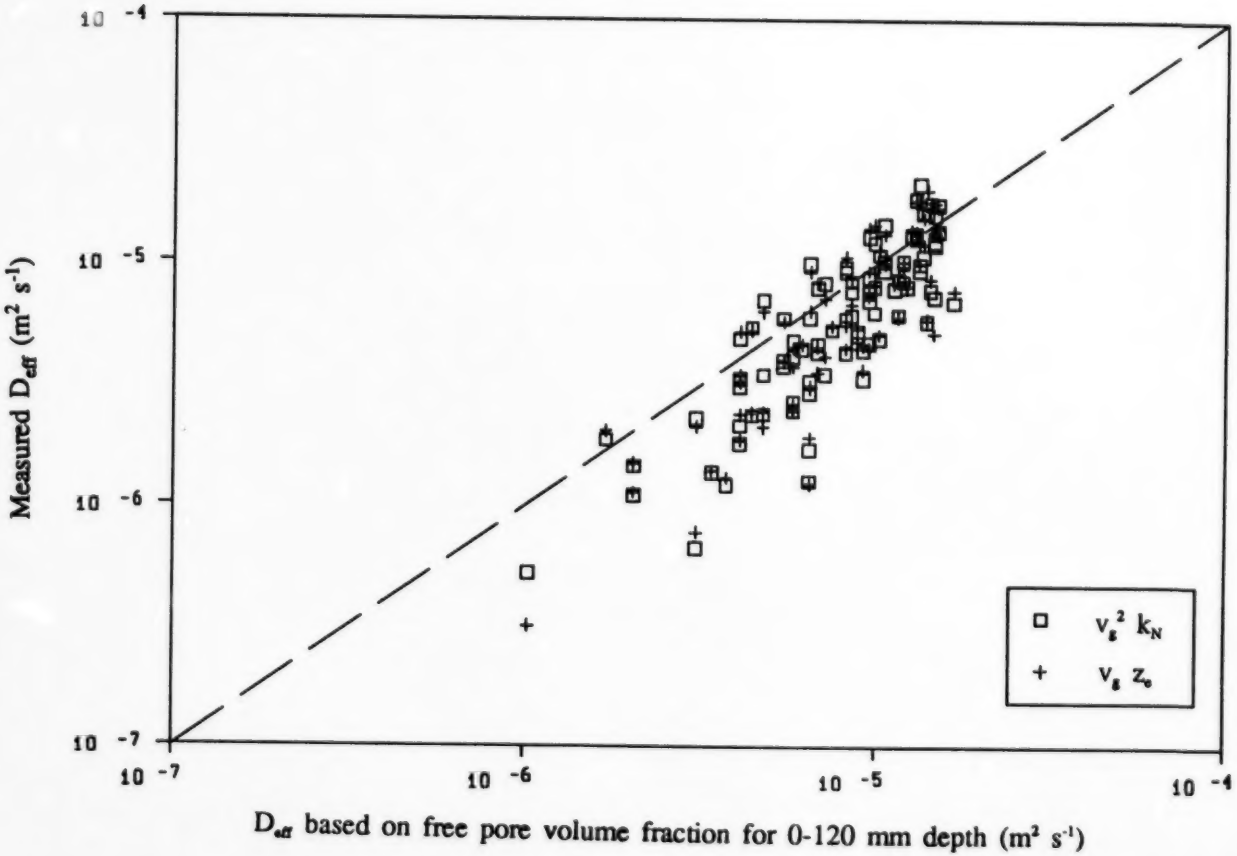

Figure 7. Comparison of effective HT soil diffusion coefficients derived from experiment to D_{eff} derived empirically from soil free pore volume and an assumed tortuosity of 1.5 (equation 13).

Table 4

Ratio of Measured Effective Soil HT Diffusion Coefficient, D_{eff}, to $D_o \theta_a/\tau$

Ratio	Mean	Standard Deviation	Geometric Mean	Geometric Standard Deviation (GSD)
$D_{eff} = v_g^2/k_N$	0.79	0.31	0.72	1.54
$D_{eff} = z_e v_g$	0.81	0.32	0.73	1.55

Little change in biological activity (<8% variation) has been observed among soil samples stored for several months (Conrad and Seiler 1981, Förstel 1986, Spencer and Dunstall 1986) indicating that the activity of the hydrogenase enzyme, responsible for HT oxidation, remained relatively constant even though the soil microflora likely underwent considerable qualitative and quantitative changes during storage. Furthermore, biological oxidation rates have been found to be independent of soil particle composition, organic content, pH, or chemical properties (McFarlane et al. 1979, Spencer and Dunstall 1986).

Accumulated experimental data therefore suggests that the availability of biological oxidation sites is of minor importance to overall variability in deposition velocity.

It therefore appears that deposition velocity is regulated usually by the processes that control the access of HT to reaction sites within the soil. These processes include the diffusion of HT into the soil (described in the theory of Appendix 1 by D_{eff}) and the diffusion of HT within the pore air to the reaction sites on pore walls (the effect of which is included in the parameter k_N). Both these processes are affected by soil water content which controls the available pore space filled with air and thus the diffusion of HT within the soil. The relationship between diffusion and free pore volume has been well documented (Currie 1961, Pritchard 1985). Soil water content would also affect the degree to which soil particles are covered by water films, thereby restricting access of the relatively insoluble HT to reaction sites located on soil particle surfaces (and again, therefore, the value of k_N).

The effect of water content on the effective HT conversion rate coefficient, k_N, was clearly observed, with values being relatively constant when the air filled volume fraction of pore space was 0.5 or greater, indicating, presumably, that no significant water film was formed and, hence, that access to reaction sites was not restricted. At higher water contents, the rate coefficient dropped steadily reaching a value approximately two orders of magnitude lower than the typical rate coefficient at air filled volume fractions greater than 0.5, suggesting that access to reaction sites was becoming restricted due to the presence of water films on soil particles with film thickness and completeness increasing with water content and/or the occlusion of pores by water lensing.

Thus both the effective diffusion coefficient of HT in soil, D_{eff}, and the effective biological oxidation rate coefficient, k_N, could be expected to decrease as soil water content increased with access to the reaction sites becoming limiting. The observed reduction in v_g with water content is due to reductions in the rates of both these processes.

Although the pore size distribution, which is dependent on the geometrical properties of soils, may also influence gaseous diffusion, we were unable to relate soil composition to variability in diffusion or deposition velocity. Organic content also affects both the amount and nature of air space in soil (Brady 1974) and may be expected to influence diffusion, although, once again, little effect of organic content (range 0.5 to 12.8% by weight) on v_g was observed.

Reductions in v_g can also occur under conditions of extremely low soil water contents, presumably by limiting the biological activity in soil. Soils with a water content <1% of soil dry weight are biologically inactive (Fallon 1982b, Conrad and Seiler 1981). Bunnenberg et al. (1986) reported a steep increase in deposition velocity as moisture content increased between 1.4 and 3.0% which was related to higher densities of soil bacteria (the change in free pore volume was minor). Thus, in extremely dry soils, HT deposition would be limited by the reduced availability of biochemical

oxidation sites. In temperate regions, however, surface soil water contents typically exceed 4% and it seems likely that reduced biological activity due to extreme soil dryness would seldom be of importance. The conversion rates of HT in the surface soil stratum would be expected to decline as a result of reduced biological activity only during prolonged dry spells.

The depth profile of deposited HTO was, in general, well approximated by an exponential decrease in concentration with depth. The exponential scaling depth, z_e, was found to vary over a narrow range (15 to 30 mm) for most soils tested. The depth of penetration is expected to increase with increasing rate of HT diffusion into the soil and decrease with increasing rate of HT oxidation. Both diffusion and effective reaction rate coefficient decrease with increasing soil water content. Our results indicate little systematic change in z_e with soil water content (as reflected by the air filled pore space volume fraction) suggesting that the effects on z_e of changes in diffusion and reaction with change in soil water content tend to counteract each other.

For the purposes of risk and impact assessment for HT handling facilities it is necessary to predict rates of oxidation of HT releases. Hence the need exists to predict HT deposition velocities to soil. In this regard, the relationship between v_g, z_e and k_N was found to be very consistent with previously developed theory (Ogram 1982). This theory forms the basis for the methods currently used to model HT deposition and the resulting HTO deposition profile within soil in the Ontario Hydro Environmental Tritium Model, ETMOD (Russell and Ogram 1988). The present results thus confirm the validity of using this method.

The theory may also provide a framework for predicting v_g on the basis of readily available information on soil conditions (eg, soil bulk density or texture and soil water content). For example, a possible way of predicting v_g is to estimate D_{eff} from a knowledge of soil bulk density and water content (using an empirical relationship like equation (13)), and then to use equation (11c) assuming a representative value of z_e, eg, 23 mm as found in the present experiments. Figure 8 shows a log-log plot of v_g values calculated in this way versus the experimental values from the present work, where it can be seen that (11c) with the above assumptions gives reasonable agreement with measurements but tends to overestimate v_g at lower v_g values. Since for low v_g's soil samples typically had high water contents, the overestimates are possibly an effect of pore-blocking, ie, a portion of the air filled pore space was not accessible by diffusion of HT from the ambient air. This problem has been recognized in the literature and a number of correlations are available to account for the effect on D_{eff}, eg, Sallam et al. (1984). Investigation of the present results in terms of these other models might provide more reliable methods of predicting v_g but is beyond the scope of present work .

We note also that measuring the rate of deposition and the deposition depth profile for a gas that reacts and is retained within the soil provides a means of determining the diffusion coefficient averaged effectively over the depth of penetration of the gas (equation (11c)). This represents a new method of determining gaseous diffusion coefficients in soil.

5.0 CONCLUSIONS

Laboratory measurements of the rate of HT deposition to eighty-eight soil samples from eleven Ontario sites have shown the following:

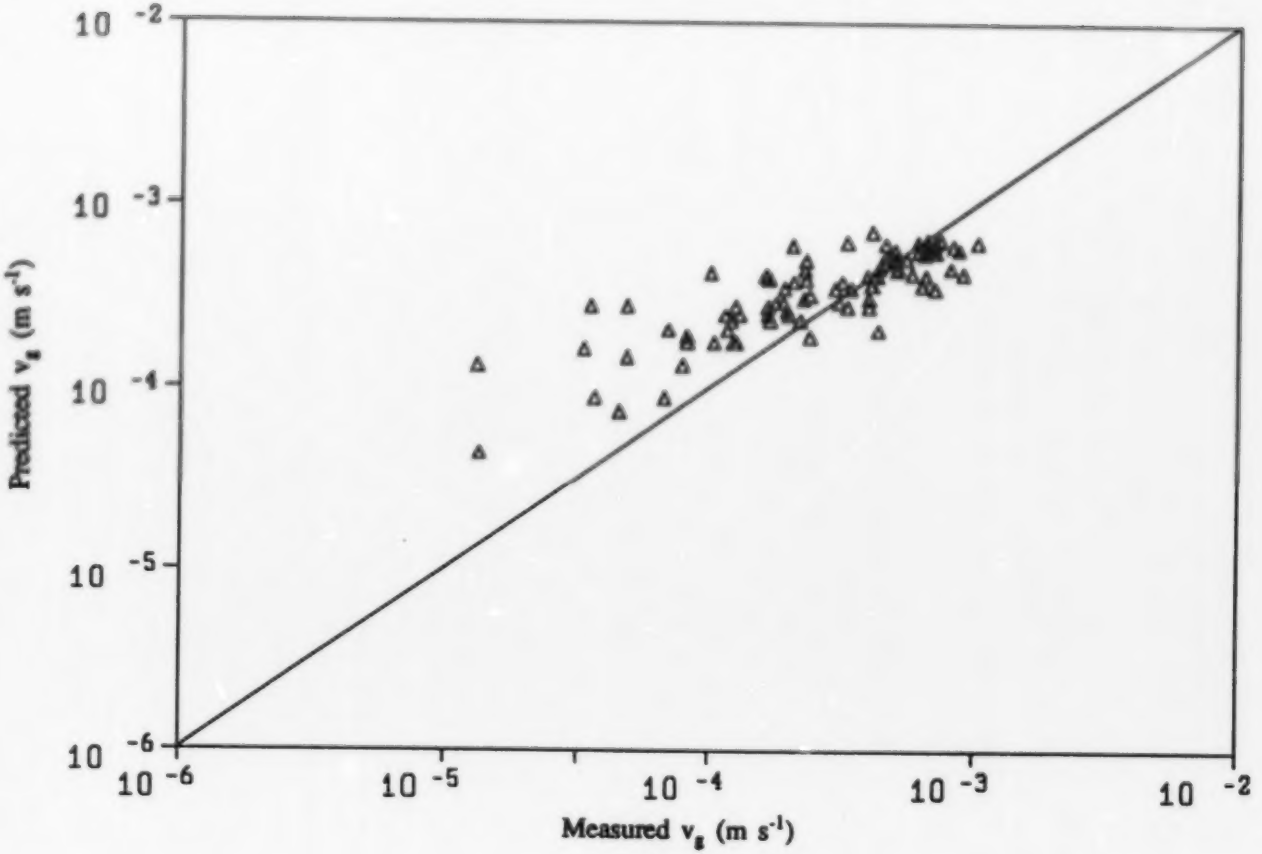

Figure 8. Comparison of predicted and measured deposition velocities. Predicted v_g's were derived from soil bulk density and water content and an assumed z_o of 23 mm.

(1) Deposition velocities varied between 0.13 x 10^{-4} and 10.2 x 10^{-4} m s^{-1}, in accord with previous laboratory and field measurements.

(2) The experiment confirmed the importance of soil water content in controlling the deposition velocity of HT to soil (particularly through its effect on diffusion of HT into the soil and access of HT to enzymatic reaction sites located on soil particles).

(3) Experimental measurements were in good agreement with the HT deposition theory developed earlier and implemented in Ontario Hydro's Environmental Tritium Model. This theory describes the deposition velocity of HT to soil in terms of the component processes of diffusion and reaction of HT in the soil.

(4) The HT deposition theory validated by the present experiments may be useful for predicting HT deposition velocities on a basis of knowledge of soil conditions alone for the purposes of impact assessment. Further work to determine appropriate methods for predicting effective diffusion coefficients for HT in soil on the basis of soil texture (or bulk density) and soil water content using the results from the present experiment is recommended.

(5) The experimental technique provides a new method of determining gaseous diffusion coefficients in soils.

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APPENDIX 1

THEORETICAL RELATIONS BETWEEN HT DEPOSITION VELOCITY TO SOIL, DEPTH OF HT PENETRATION INTO SOIL, EFFECTIVE HT DIFFUSION COEFFICIENT IN SOIL AND RATE OF OXIDATION OF HT IN SOIL

Nomenclature

θ_p	Soil porosity (fraction of total soil volume)
θ_w	Soil water content (fraction of total soil volume)
$\theta_a = \theta_p - \theta_w$	Free (air filled) pore volume (fraction of total soil volume)
[HT] [Bq m^{-3}]:	Concentration of HT in air above soil
$[HT]_z$ [Bq m^{-3}]:	Concentration of HT in soil (per unit volume of air)
$[HTO]_z$ [Bq m^{-3}]:	Concentration of HTO in soil (per unit volume of soil)
J_z [Bq m^{-2} s^{-1}]:	HT flux into soil (per unit horizontal soil area)
v_g [m s^{-1}]:	Deposition velocity of HT to soil.
D_{eff} [m^2 s^{-1}]:	Effective diffusion coefficient for HT in soil
k [s^{-1}]:	Rate coefficient for oxidation of HT in soil
k_N (θ_a k) [s^{-1}]:	Rate coefficient for formation of HTO in soil

1. Derivation of Relations

The purpose of this Appendix is to summarize the derivation of the relationships between v_g, k (or k_N), z_e and D_{eff} given previously [Ogram 1982, 1984; Russell and Ogram 1988]. The situation modelled is shown schematically in Figure A-1, where we consider a soil surface exposed to a constant concentration of HT in air for a duration, Δt. The HT diffuses into the soil where it is converted to HTO by microbial action.

If it is assumed that HT is dilute in soil air and that mass flow is negligible then Fick's law of diffusion [Crank 1975; Cussler 1984] can be used to describe HT movement in soil air. An effective soil diffusion coefficient, D_{eff}, can be defined [Currie 1961, Cussler 1984] as follows:

$$J_z = - D_{eff} \frac{\partial [HT]_z}{\partial z} \qquad (1\text{-}1)$$

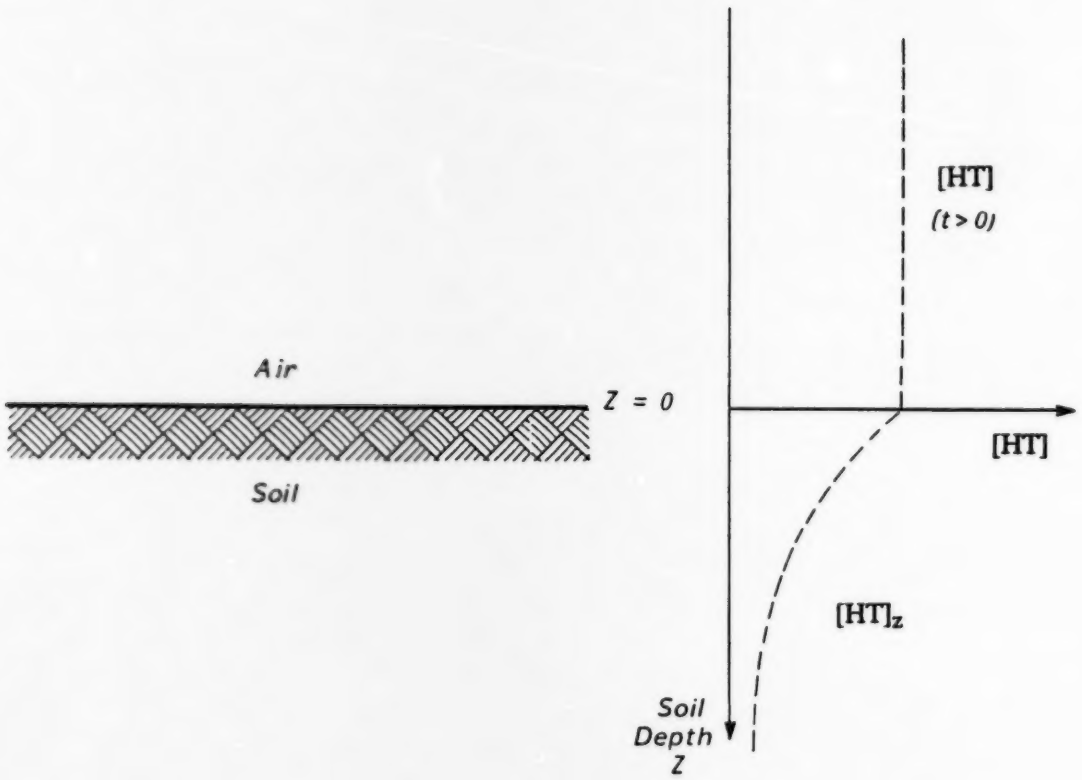

Figure A-1. Schematic of HT profile in air above soil and within soil pore air.

D_{eff} takes into account the effect of limited porosity and the tortuosity of the air path that HT must follow in order to diffuse into the soil.

As HT reacts within the soil, $[HT]_z$ is described by the one-dimensional (z) continuity equation appropriate for a species that diffuses into and reacts within a horizontally homogeneous medium:

$$\frac{\theta_a \, \partial [HT]_z}{\partial t} = D_{eff} \frac{\partial^2 [HT]_z}{\partial z^2} - k \, \theta_a \, [HT]_z$$

or

$$\frac{\partial [HT]_z}{\partial t} = (D_{eff} / \theta_a) \frac{\partial^2 [HT]_z}{\partial z^2} - k \, [HT]_z \qquad (1\text{-}2)$$

where the factor θ_a accounts for the fact that the flux J_z is expressed per unit area of soil whereas $[HT]_z$ is expressed per unit volume of soil air: ie, the concentration of HT per unit volume of soil is $\theta_a \, [HT]_z$. The continuity equation (1-2) can be derived by considering the diffusive flux into and out of a reference volume and the production and loss of the species of interest within that volume [Crank 1975; Cussler 1984]. Soil properties are assumed to be unchanging over the depth of significant penetration of HT into the soil (ie, D_{eff} and k_N are assumed to be constant and independent of depth) and the rate of conversion of HT has been assumed to be first order (which seems reasonable as HT concentrations are extremely low; in the environment HT will likely always be a small fraction of ambient H_2).

Steady state corresponds to an $[HT]_z$ that is unchanging in time, ie, the left hand side of (1-2) approaches zero. On assuming steady state and applying the boundary condition that the concentration of HT in soil air is equal to the concentration in air above the surface (z = 0), ie,

$$[HT]_z(z=0) = [HT], \qquad (1\text{-}3)$$

equation (1-2) can be solved to yield

$$[HT]_z = [HT] \exp\{-z \, (k \, \theta_a / D_{eff})^{0.5}\} \qquad (1\text{-}4)$$

or

$$[HT]_z = [HT] \exp\{-z / z_e\}. \qquad (1\text{-}5)$$

This solution predicts that $[HT]_z$ exhibits an exponential profile with a scaling depth,

$$z_e = (D_{eff} \, (k \, \theta_a)^{-1})^{0.5}.$$

$$= (D_{eff} / k_N)^{0.5} \qquad (1\text{-}6)$$

The Ontario Hydro code, ETMOD, considers a boundary to HT diffusion at a finite soil depth, L. The solution (as used in ETMOD; Russell and Ogram 1988) reduces to the above for $z_e << L$.

By definition, the deposition flux of HT to the soil surface is given by the product of the deposition velocity and the air concentration and is equal to $J_Z(z=0)$. The reference height for deposition velocity is usually 1 m above the surface. We now assume that the resistance to surface uptake is the rate limiting step for HT deposition to soils. It follows that the gradient in [HT] in the air to 1 m is small, ie, [HT] at 1 m is approximately equal to that immediately above the surface, so that

$$J_Z(z=0) = v_g\ [HT]. \tag{1-7}$$

From (1-1), (1-4) and (1-7) we obtain

$$\begin{aligned} v_g = J_Z(z=0) / [HT] &= -(D_{eff} / [HT]) \left. \frac{\partial [HT]_Z}{\partial z} \right|_{z=0} \\ &= (\theta_a\ k\ D_{eff})^{0.5} \\ &= (k_N\ D_{eff})^{0.5} \end{aligned} \tag{1-8}$$

The following relations can also be derived,

$$v_g = k_N\ z_e \tag{1-9}$$

$$= D_{eff} / z_e \tag{1-10}$$

If the movement of HTO is small over the exposure period, Δt, then the profile of the deposited HTO resulting from exposure should remain similar to that of the HT, with the concentration of HTO per unit volume of soil at depth z being given by

$$\begin{aligned} [HTO]_Z &= \theta_a\ k\ [HT]_Z\ \Delta t \\ &= k_N\ [HT]_Z\ \Delta t \end{aligned} \tag{1-8}$$

so that k_N is a rate coefficient for the formation of HTO. For short enough exposures the initial HTO deposit should exhibit an exponential profile similar to that of the HT (1-5).

2. Justification of Assumptions

For the above theory to apply a number of assumptions must be shown to be valid. We examine briefly the validity of these assumptions for the experiments reported here.

(1) In order for the HT profile in soil to be near-steady state, the characteristic time for diffusion of HT in the soil over the depth of the profile must be less than the duration of the test, Δt. Figure A-2 (Ogram 1984) shows the evolution of the depth profile of non-dimensional HT concentration in soil, $[HT]_z / [HT]$, against non-dimensional depth, z/z_e, at various non-dimensional times, $\tau = t\ D_{eff} / z_e^2$. These profiles were obtained from the full time dependent solution to the continuity equation (1-3). At the non-dimensional time $\tau = 1$, the profile is close to the steady state solution, given by (1-5).

Noting that $\tau = t\ D_{eff}/z_e^2 = t\ v_g/z_e$, we can calculate representative values of $t_1 = z_e / v_g$, corresponding to $\tau = 1$, and compare them to the duration of our experiments, Δt, to check if steady-state was achieved. Table A-1 shows values of t_1 calculated for various v_g and z_e values typical of our experiments. In most cases z_e was about 20 to 30 mm, with v_g values between 1 and 10 x 10^{-4} m s^{-1}, so that values of t_1 would range between fractions of a minute to a few minutes, which can be compared to exposure times, Δt, of 26 to 49 min. Equilibrium would thus have been achieved in the present experiments, except at very low v_g.

TABLE A-1

Time t_1 (min.) to Reach a Near Steady-State Soil HT Depth Profile

v_g x 10^4 (m s^{-1})	z_e x 10^2 (m) (m)	t_1 (min)
0.1	1	17
	10	170
1	1	1.7
	10	17
10	1	0.17
	10	1.7

(2) For the HTO profile at the end of the experiment to approximate the steady state HT profile, the characteristic time for diffusion of HTO, equal to $z_e^2 / D_{eff}(HTO)$, must be much less than the exposure time, Δt. The effective diffusion coefficient for HTO in soil is very small (~ 10^{-9} m^2 s^{-1}, Garland 1980) due to almost all the HTO being present in the liquid phase. For a typical z_e of 0.03 m (30 mm) this gives a time constant of about 250 h, which is much longer than the exposure time (less than 1h); the assumption therefore holds.

(3) For [HT] at 1-m height to be approximately equal to that at the soil surface the deposition of HT to soils must be surface uptake limited. The deposition velocity can be written as $v_g = (R_a + R_s)^{-1}$ where R_s is the resistance to uptake at the surface and R_a is the resistance to atmospheric transfer. R_a includes resistance to turbulent transport from the reference height (1 m) to the surface laminar sublayer, and diffusive transport through the laminar sublayer. If uptake is rate limiting then $v_g \sim R_s^{-1}$. Typically $R_a \sim 10^{-2}$ m s^{-1}, thus for the HT deposition velocities measured here, which were typically one or two orders of magnitude lower, surface uptake was probably limiting, ie, $v_g << 0.01$ m s^{-1}.

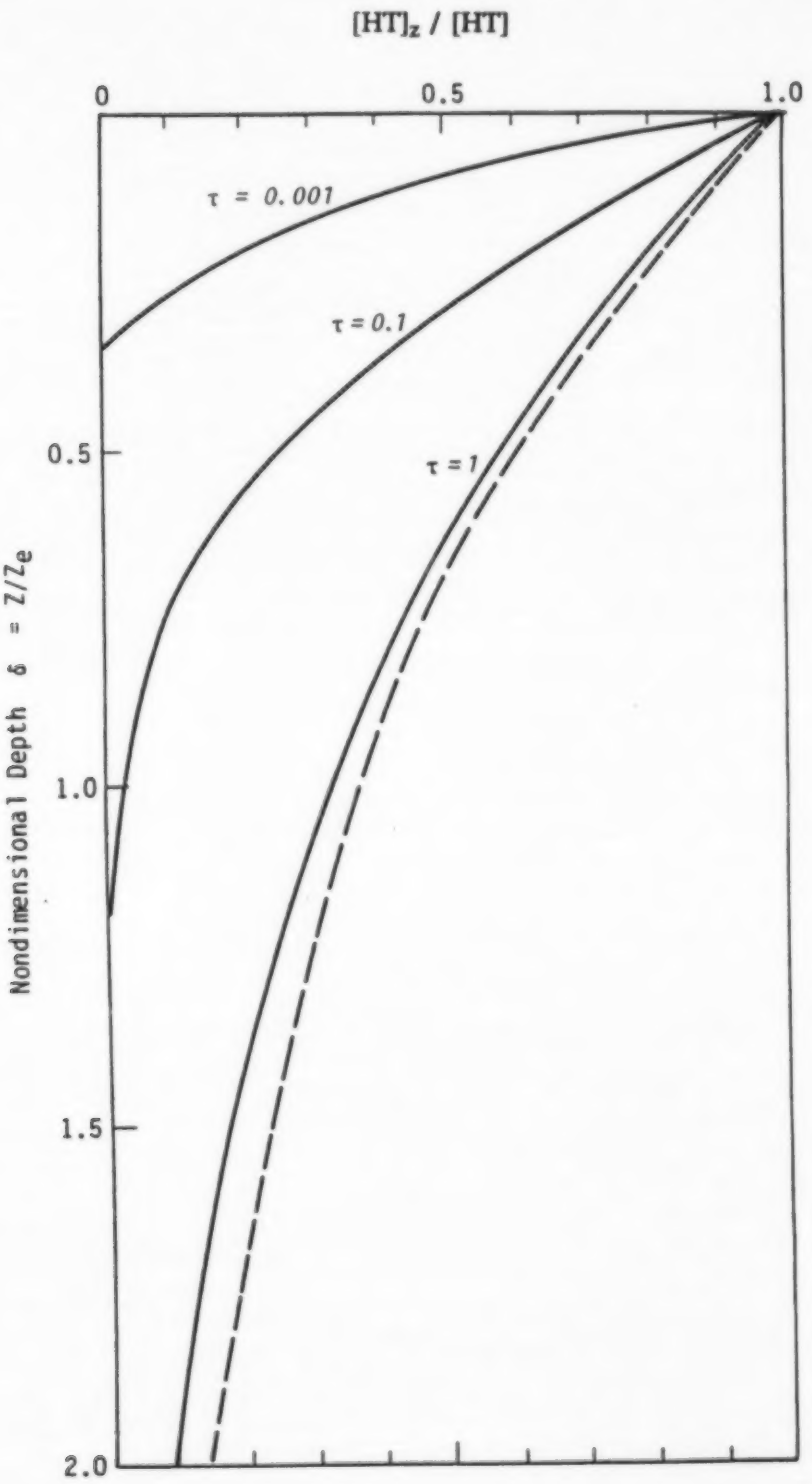

Figure A-2. Normalized HT concentration profiles calculated for various non-dimensional times, τ . The dashed line shows the steady state profile obtained as τ becomes large (from Ogram 1984).

APPENDIX 2

Summary of Data - HT Deposition to Eleven Ontario Soils

Site	Code	Soil Treatment*		Vg (m/s)	kN (1/s)	Ze*1E2 (m)	Bulk Density*1E3 (kg/m^3) (0-15mm)	Bulk Density*1E3 (kg/m^3) (0-120mm)	%H20 of dry wt. (0-15mm)	%H20 of dry wt. (0-120mm)	%Organic (by wt.)
CHERRYWOOD TS	C	A	1	6.08E-04	2.78E-02	2.16	0.80	0.71	54.10	49.60	10.50
	C	A	2	5.03E-04	2.34E-02	2.26	0.80	0.80	56.70	49.80	10.50
	C	D	1	6.91E-04	2.60E-02	2.61	0.80	0.71	53.00	49.10	10.50
	C	D	2	5.47E-04	2.93E-02	1.86	1.00	0.78	47.90	46.40	10.50
	C	X	1	2.03E-04	7.18E-03	2.88	0.60	0.60	70.10	60.80	10.50
	C	X	2	2.27E-04	1.06E-02	2.23	0.60	0.73	69.10	59.30	10.50
	C	2X	1	1.67E-04	7.06E-03	2.37	0.50	0.79	77.40	68.30	10.50
	C	2X	2	1.29E-04	6.23E-03	2.02	0.60	0.78	83.60	69.70	10.50
WESLEYVILLE	W	A	1	3.28E-04	1.81E-02	1.93	1.20	1.34	26.40	23.10	5.20
	W	A	2	2.37E-04	1.04E-02	2.22	1.70	1.36	28.20	25.80	5.20
	W	D	1	3.94E-04	1.56E-02	2.33	1.40	1.32	23.80	23.10	5.20
	W	D	2	4.29E-04	2.64E-02	1.47	1.80	1.45	22.90	21.80	5.20
	W	X	1	6.69E-05	3.09E-03	2.22	1.10	1.36	36.30	31.30	5.20
	W	X	2	4.84E-05	1.71E-03	2.86	1.20	1.35	35.40	29.10	5.20
	W	2X	1	6.92E-05	1.39E-03	3.03	1.60	1.26	34.40	30.00	5.20
	W	2X	2	1.33E-05	3.40E-04	2.35	2.00	1.40	37.90	31.60	5.20
DARLINGTON	D	A	1	6.53E-04	3.15E-02	2.13	0.70	1.03	17.00	17.10	6.60
	D	A	2	8.64E-04	3.52E-02	2.11	1.00	1.10	19.00	17.70	6.60
	D	D	1	6.45E-04	4.20E-02	1.88	1.10	1.16	18.20	14.90	6.60
	D	D	2	6.54E-04	4.01E-02	2.36	1.20	1.12	13.80	15.50	6.60
	D	X	1	4.94E-04	2.90E-02	1.92	1.10	1.16	27.10	18.10	6.60
	D	X	2	4.05E-04	2.38E-02	1.90	1.10	0.85	27.10	21.80	6.60
	D	2X	1	3.27E-04	1.48E-02	1.56	1.20	0.88	30.30	27.70	6.60
	D	2X	2	2.96E-04	9.49E-03	1.87	1.40	1.23	28.00	24.10	6.60
PICKERING	P	A	1	6.04E-04	3.10E-02	2.23	0.50	0.98	26.10	20.60	9.80
	P	A	2	8.22E-04	4.18E-02	2.44	0.50	0.86	39.50	31.10	9.80
	P	D	1	8.05E-04	4.48E-02	1.63	0.70	1.03	42.50	29.90	9.80
	P	D	2	6.69E-04	2.80E-02	1.77	0.60	0.99	23.50	23.40	9.80
	P	X	1	4.13E-04	2.25E-02	1.76	0.70	0.90	54.80	42.20	9.80
	P	X	2	3.94E-04	1.88E-02	1.81	0.80	1.02	53.70	39.20	9.80
	P	2X	1	2.24E-04	1.45E-02	1.84	0.80	0.95	58.30	45.50	9.80
	P	2X	2	2.17E-04	7.98E-03	2.63	1.00	0.99	59.90	47.40	9.80

APPENDIX 2 (cont'd)

Summary of Data - HT Deposition to Eleven Ontario Soils

Site	Code	Soil Treatment*		Vg (m/s)	kN (1/s)	Ze*1E2 (m)	Bulk Density*1E3 (kg/m^3) (0-15mm)	(0-120mm)	%H20 of dry wt. (0-15mm)	(0-120mm)	%Organic (by wt.)
N. BOWMANVILLE-ORG	O	A	1	1.03E-04	3.20E-03	3.33	0.90	1.19	36.60	36.40	5.30
	O	A	2	1.20E-04	3.86E-03	3.24	1.20	1.22	30.90	30.50	5.30
	O	D	1	1.23E-04	3.12E-03	4.22	0.80	1.10	33.90	42.20	5.30
	O	D	2	9.99E-05	1.60E-03	8.13	0.50	0.89	49.80	42.20	5.30
	O	X	1	4.51E-05	1.09E-03	4.48	0.70	1.24	43.80	39.20	5.30
	O	X	2	3.64E-05	1.21E-03	3.10	0.90	1.00	41.80	56.70	5.30
	O	2X	1	3.50E-05	7.22E-04	5.43	0.70	0.96	42.90	46.70	5.30
	O	2X	2	3.31E-05	9.03E-04	3.92	0.80	0.99	48.50	52.10	5.30
N BOWMANVILLE-SAND	S	A	1	7.02E-04	5.12E-02	1.51	1.30	1.54	18.00	11.90	0.50
	S	A	2	5.74E-04	4.13E-02	1.61	1.30	1.46	13.90	11.00	0.50
	S	D	1	9.00E-04	6.69E-02	1.61	1.20	1.50	12.70	9.62	0.50
	S	D	2	6.29E-04	5.21E-02	1.38	1.40	1.65	10.20	7.66	0.50
	S	X	1	1.16E-04	5.68E-03	2.09	1.50	1.61	21.00	15.70	0.50
	S	X	2	8.13E-05	2.82E-03	2.97	1.40	1.59	20.80	17.20	0.50
	S	2X	1	7.85E-05	2.69E-03	2.69	1.20	1.50	30.60	22.60	0.50
	S	2X	2	1.24E-04	5.31E-03	2.51	1.10	1.39	29.00	20.30	0.50
BOLTON	B	A	1	2.37E-04	1.06E-02	2.31	0.70	0.95	49.30	43.90	10.50
	B	A	2	3.04E-04	1.17E-02	2.72	0.70	0.98	48.00	44.30	10.50
	B	D	1	4.31E-04	1.99E-02	2.31	0.70	0.89	34.70	39.80	10.50
	B	D	2	4.05E-04	1.95E-02	2.20	0.80	0.98	34.50	39.60	10.50
	B	X	1	1.25E-04	7.33E-03	1.89	0.90	1.04	53.30	47.30	10.50
	B	X	2	1.97E-04	9.47E-03	2.26	0.60	0.89	64.90	55.90	10.50
	B	2X	1	1.31E-05	2.59E-04	5.88	0.70	0.98	69.00	55.70	10.50
	B	2X	2	1.94E-04	8.53E-03	2.38	0.80	0.89	63.80	54.20	10.50
CLAREMONT	L	A	1	3.37E-04	1.93E-02	1.72	1.20	0.91	43.90	45.10	12.80
	L	A	2	4.01E-04	2.65E-02	1.68	0.60	0.91	47.90	44.40	12.80
	L	D	1	4.93E-04	2.83E-02	1.81	1.00	0.81	36.40	43.00	12.80
	L	D	2	4.94E-04	2.62E-02	2.03	0.90	0.81	30.30	37.40	12.80
	L	X	1	1.76E-04	7.30E-03	2.01	1.00	0.86	61.10	55.50	12.80
	L	X	2	1.89E-04	8.36E-03	2.35	0.60	0.79	70.50	58.60	12.80
	L	2X	1	1.64E-04	5.64E-03	2.27	1.00	0.80	78.40	65.50	12.80
	L	2X	2	1.13E-04	5.16E-03	2.17	0.90	0.81	71.80	64.30	12.80

APPENDIX 2 (cont'd)

Summary of Data - HT Deposition to Eleven Ontario Soils

Site	Code	Soil Treatment*		Vg (m/s)	kN (1/s)	Ze*1E2 (m)	Bulk Density*1E3 (kg/m^3) (0-15mm)	Bulk Density*1E3 (kg/m^3) (0-120mm)	%H20 of dry wt. (0-15mm)	%H20 of dry wt. (0-120mm)	%Organic (by wt.)
GLEN MAJOR	G	A	1	4.54E-04	2.67E-02	1.88	0.80	0.63	61.50	55.70	11.10
	G	A	2	4.55E-04	2.60E-02	1.77	1.00	0.78	46.30	44.30	11.10
	G	D	1	6.43E-04	3.16E-02	2.12	0.90	0.83	31.70	38.00	11.10
	G	D	2	7.35E-04	3.11E-02	2.31	1.10	0.78	27.90	34.80	11.10
	G	X	1	1.61E-04	5.57E-03	2.77	0.90	0.78	60.90	55.80	11.10
	G	X	2	2.28E-04	8.50E-03	2.63	0.90	0.75	58.00	50.80	11.10
	G	2X	1	1.87E-04	7.57E-03	2.40	1.00	0.80	55.70	62.60	11.10
	G	2X	2	1.22E-04	4.85E-03	2.63	0.80	0.80	71.10	72.50	11.10
BALLANTRAE	A	A	1	1.02E-03	6.60E-02	1.74	1.00	1.09	18.40	14.80	3.50
	A	A	2	6.51E-04	2.41E-02	2.75	1.00	1.11	15.70	14.50	3.50
	A	D	1	7.04E-04	4.04E-02	1.89	0.80	1.07	21.60	15.60	3.50
	A	D	2	7.04E-04	3.92E-02	1.93	1.00	1.20	13.60	14.20	3.50
	A	X	1	4.53E-04	2.63E-02	1.83	0.90	1.14	27.90	21.50	3.50
	A	X	2	4.23E-04	2.06E-02	2.19	0.90	1.15	28.90	25.00	3.50
	A	2X	1	1.66E-04	8.28E-03	2.18	0.90	1.06	37.80	31.10	3.50
	A	2X	2	2.06E-04	9.08E-03	2.31	1.30	1.11	36.40	28.70	3.50
KINSALE	K	A	1	3.89E-04	2.17E-02	1.95	0.90	0.93	47.50	40.60	9.00
	K	A	2	6.47E-04	3.28E-02	2.16	0.60	0.91	50.00	41.40	9.00
	K	D	1	3.13E-04	1.87E-02	1.71	1.20	1.05	36.40	33.00	9.00
	K	D	2	5.08E-04	2.54E-02	2.05	1.10	0.94	37.10	36.30	9.00
	K	X	1	2.29E-04	1.20E-02	2.00	0.80	0.86	50.00	46.80	9.00
	K	X	2	1.64E-04	8.28E-03	1.86	1.20	0.95	51.50	47.70	9.00
	K	2X	1	4.83E-05	1.86E-03	2.53	1.10	0.89	60.10	53.50	9.00
	K	2X	2	8.13E-05	3.68E-03	2.27	1.20	1.01	53.50	49.40	9.00

* A - Natural
B - Dried
X - Water Added (Amount depended on soil type)
2X - Double Amount Water Added

Appendix 3a

Derived Soil HT Deposition Velocities (m s^{-1})

Site	Code	Soil Treatment*		Vg (meas)	Vg SQR(kN*D)	Vg kN*Ze	Vg Deff/Ze	Vg Deff/ 0.023
CHERRYWOOD TS	C	A	1	6.08E-04	5.99E-04	6.00E-04	5.98E-04	5.62E-04
	C	A	2	5.03E-04	4.89E-04	5.28E-04	4.52E-04	4.43E-04
	C	D	1	6.91E-04	5.80E-04	6.79E-04	4.95E-04	5.62E-04
	C	D	2	5.47E-04	5.90E-04	5.46E-04	6.39E-04	5.17E-04
	C	X	1	2.03E-04	3.16E-04	2.07E-04	4.84E-04	6.06E-04
	C	X	2	2.27E-04	3.29E-04	2.36E-04	4.58E-04	4.43E-04
	C	2X	1	1.67E-04	1.96E-04	1.67E-04	2.30E-04	2.37E-04
	C	2X	2	1.29E-04	1.90E-04	1.26E-04	2.86E-04	2.51E-04
WESLEYVILLE	W	A	1	3.28E-04	3.42E-04	3.49E-04	3.35E-04	2.81E-04
	W	A	2	2.37E-04	2.14E-04	2.31E-04	1.99E-04	1.92E-04
	W	D	1	3.94E-04	3.17E-04	3.64E-04	2.77E-04	2.81E-04
	W	D	2	4.29E-04	3.54E-04	3.87E-04	3.25E-04	2.07E-04
	W	X	1	6.69E-05	7.94E-05	6.87E-05	9.18E-05	8.87E-05
	W	X	2	4.84E-05	7.62E-05	4.89E-05	1.19E-04	1.48E-04
	W	2X	1	6.92E-05	8.13E-05	4.21E-05	1.57E-04	2.07E-04
	W	2X	2	1.33E-05	1.86E-05	8.00E-06	4.34E-05	4.43E-05
DARLINGTON	D	A	1	6.53E-04	6.86E-04	6.72E-04	7.02E-04	6.50E-04
	D	A	2	8.64E-04	6.83E-04	7.41E-04	6.30E-04	5.77E-04
	D	D	1	6.45E-04	7.46E-04	7.89E-04	7.05E-04	5.77E-04
	D	D	2	6.54E-04	7.38E-04	9.46E-04	5.77E-04	5.91E-04
	D	X	1	4.94E-04	5.87E-04	5.58E-04	6.19E-04	5.17E-04
	D	X	2	4.05E-04	6.30E-04	4.52E-04	8.76E-04	7.24E-04
	D	2X	1	3.27E-04	4.65E-04	2.31E-04	9.36E-04	6.36E-04
	D	2X	2	2.96E-04	2.78E-04	1.78E-04	4.36E-04	3.55E-04
PICKERING	P	A	1	6.04E-04	6.73E-04	6.90E-04	6.56E-04	6.36E-04
	P	A	2	8.22E-04	7.63E-04	1.02E-03	5.72E-04	6.06E-04
	P	D	1	8.05E-04	6.87E-04	7.31E-04	6.46E-04	4.58E-04
	P	D	2	6.69E-04	6.17E-04	4.96E-04	7.67E-04	5.91E-04
	P	X	1	4.13E-04	4.63E-04	3.96E-04	5.41E-04	4.14E-04
	P	X	2	3.94E-04	3.66E-04	3.41E-04	3.94E-04	3.10E-04
	P	2X	1	2.24E-04	3.22E-04	2.67E-04	3.88E-04	3.10E-04
	P	2X	2	2.17E-04	2.08E-04	2.10E-04	2.07E-04	2.37E-04
N. BOWMANVILLE-ORG	O	A	1	1.03E-04	1.14E-04	1.07E-04	1.22E-04	1.77E-04
	O	A	2	1.20E-04	1.45E-04	1.25E-04	1.68E-04	2.37E-04
	O	D	1	1.23E-04	1.13E-04	1.32E-04	9.67E-05	1.77E-04
	O	D	2	9.99E-05	1.26E-04	1.30E-04	1.21E-04	4.29E-04
	O	X	1	4.51E-05	4.30E-05	4.89E-05	3.79E-05	7.39E-05
	O	X	2	3.64E-05	4.97E-05	3.75E-05	6.59E-05	8.87E-05
	O	2X	1	3.50E-05	6.83E-05	3.92E-05	1.19E-04	2.81E-04
	O	2X	2	3.31E-05	5.81E-05	3.54E-05	9.54E-05	1.63E-04
N BOWMANVILLE-SAND	S	A	1	7.02E-04	6.46E-04	7.75E-04	5.39E-04	3.55E-04
	S	A	2	5.74E-04	6.38E-04	6.64E-04	6.13E-04	4.29E-04
	S	D	1	9.00E-04	8.12E-04	1.07E-03	6.14E-04	4.29E-04
	S	D	2	6.29E-04	6.65E-04	7.17E-04	6.18E-04	3.70E-04
	S	X	1	1.16E-04	1.64E-04	1.19E-04	2.28E-04	2.07E-04
	S	X	2	8.13E-05	1.12E-04	8.37E-05	1.49E-04	1.92E-04
	S	2X	1	7.85E-05	9.07E-05	7.23E-05	1.14E-04	1.33E-04
	S	2X	2	1.24E-04	1.85E-04	1.33E-04	2.58E-04	2.81E-04
BOLTON	B	A	1	2.37E-04	2.82E-04	2.45E-04	3.23E-04	3.25E-04
	B	A	2	3.04E-04	2.82E-04	3.19E-04	2.50E-04	2.96E-04
	B	D	1	4.31E-04	4.58E-04	4.61E-04	4.55E-04	4.58E-04
	B	D	2	4.05E-04	4.07E-04	4.29E-04	3.87E-04	3.70E-04
	B	X	1	1.25E-04	1.73E-04	1.39E-04	2.16E-04	1.77E-04
	B	X	2	1.97E-04	2.34E-04	2.14E-04	2.56E-04	2.51E-04
	B	2X	1	1.31E-05	2.82E-05	1.52E-05	5.20E-05	1.33E-04
	B	2X	2	1.94E-04	2.28E-04	2.03E-04	2.58E-04	2.66E-04

Appendix 3a (cont'd)

Derived Soil HT Deposition Velocities ($m\ s^{-1}$)

Site	Code	Soil Treatment*		Vg (meas)	Vg SQR(kN*D)	Vg kN*Ze	Vg Deff/Ze	Vg Deff/ 0.023
CLAREMONT	L	A	1	3.37E-04	3.97E-04	3.31E-04	4.76E-04	3.55E-04
	L	A	2	4.01E-04	4.75E-04	4.45E-04	5.07E-04	3.70E-04
	L	D	1	4.93E-04	5.72E-04	5.13E-04	6.38E-04	5.03E-04
	L	D	2	4.94E-04	5.89E-04	5.31E-04	6.54E-04	5.77E-04
	L	X	1	1.76E-04	2.23E-04	1.47E-04	3.38E-04	2.96E-04
	L	X	2	1.89E-04	2.61E-04	1.96E-04	3.48E-04	3.55E-04
	L	2X	1	1.64E-04	1.81E-04	1.28E-04	2.55E-04	2.51E-04
	L	2X	2	1.13E-04	1.73E-04	1.12E-04	2.66E-04	2.51E-04
GLEN MAJOR	G	A	1	4.54E-04	6.17E-04	5.02E-04	7.60E-04	6.21E-04
	G	A	2	4.55E-04	5.64E-04	4.59E-04	6.93E-04	5.32E-04
	G	D	1	6.43E-04	6.30E-04	6.71E-04	5.93E-04	5.47E-04
	G	D	2	7.35E-04	6.82E-04	7.20E-04	6.46E-04	6.50E-04
	G	X	1	1.61E-04	2.30E-04	1.54E-04	3.44E-04	4.14E-04
	G	X	2	2.28E-04	3.13E-04	2.24E-04	4.39E-04	5.03E-04
	G	2X	1	1.87E-04	2.27E-04	1.82E-04	2.83E-04	2.96E-04
	G	2X	2	1.22E-04	1.41E-04	1.28E-04	1.55E-04	1.77E-04
BALLANTRAE	A	A	1	1.02E-03	9.82E-04	1.15E-03	8.42E-04	6.36E-04
	A	A	2	6.51E-04	5.87E-04	6.62E-04	5.20E-04	6.21E-04
	A	D	1	7.04E-04	7.69E-04	7.64E-04	7.73E-04	6.36E-04
	A	D	2	7.04E-04	7.12E-04	7.57E-04	6.69E-04	5.62E-04
	A	X	1	4.53E-04	5.43E-04	4.81E-04	6.14E-04	4.88E-04
	A	X	2	4.23E-04	4.43E-04	4.51E-04	4.35E-04	4.14E-04
	A	2X	1	1.66E-04	2.76E-04	1.81E-04	4.20E-04	3.99E-04
	A	2X	2	2.06E-04	2.83E-04	2.10E-04	3.83E-04	3.84E-04
KINSALE	K	A	1	3.89E-04	4.55E-04	4.23E-04	4.88E-04	4.14E-04
	K	A	2	6.47E-04	5.59E-04	7.10E-04	4.40E-04	4.14E-04
	K	D	1	3.13E-04	4.07E-04	3.20E-04	5.16E-04	3.84E-04
	K	D	2	5.08E-04	5.17E-04	5.20E-04	5.14E-04	4.58E-04
	K	X	1	2.29E-04	3.32E-04	2.40E-04	4.59E-04	3.99E-04
	K	X	2	1.64E-04	2.31E-04	1.54E-04	3.48E-04	2.81E-04
	K	2X	1	4.83E-05	1.10E-04	4.71E-05	2.55E-04	2.81E-04
	K	2X	2	8.13E-05	1.23E-04	8.36E-05	1.80E-04	1.77E-04

* A - Natural
D - Air Dried
X - Water Added (Amount depended on soil type.)
2X - Double Amount Water Added

Appendix 3b

Derived HT Oxidation Rate k, Soil Porosities and Fraction of Pore Space with Air and Water

Site	Code	Soil Treatment	Replicate	kH (1/s)	k=kH/Ap (1/s)	Ze*1E2 (m)	Deff*1E4 [Do*AP/T] (m^2/s)	Deff*1E4 [Vg2/kH] (m^2/s)	Deff*1E4 [vg*Ze] (m^2/s)	BD*1E3 (kg/m^3)	%H2O	%Pore Spe	Theta W	Theta A	FPH2O	FPair
CHERRYWOOD TS	C	A	1	2.78E-02	7.32E-02	2.16	0.13	0.13	0.13	0.71	49.60	0.73	0.35	0.38	0.48	0.52
	C	A	2	2.34E-02	7.80E-02	2.26	0.10	0.11	0.11	0.80	49.80	0.70	0.40	0.30	0.57	0.43
	C	D	1	2.60E-02	6.84E-02	2.61	0.13	0.18	0.18	0.71	49.10	0.73	0.35	0.38	0.48	0.52
	C	D	2	2.93E-02	8.37E-02	1.86	0.12	0.10	0.10	0.78	46.40	0.71	0.36	0.35	0.51	0.49
	C	X	1	7.18E-03	1.75E-02	2.88	0.14	0.06	0.06	0.68	60.80	0.77	0.36	0.41	0.47	0.53
	C	X	2	1.06E-02	3.53E-02	2.23	0.10	0.05	0.05	0.73	59.30	0.73	0.43	0.30	0.59	0.41
	C	2X	1	7.06E-03	4.41E-02	2.37	0.05	0.04	0.04	0.79	68.30	0.70	0.54	0.16	0.77	0.23
	C	2X	2	6.23E-03	3.66E-02	2.02	0.06	0.03	0.03	0.78	69.70	0.71	0.54	0.17	0.76	0.24
WESLEYVILLE	W	A	1	1.81E-02	9.53E-02	1.93	0.06	0.06	0.06	1.34	23.10	0.50	0.31	0.19	0.62	0.38
	W	A	2	1.04E-02	8.00E-02	2.22	0.04	0.05	0.05	1.36	25.80	0.49	0.35	0.13	0.72	0.28
	W	D	1	1.56E-02	8.21E-02	2.33	0.06	0.10	0.09	1.32	23.10	0.50	0.31	0.19	0.61	0.39
	W	D	2	2.64E-02	1.89E-01	1.47	0.05	0.07	0.06	1.45	21.80	0.45	0.32	0.14	0.70	0.30
	W	X	1	3.09E-03	5.15E-02	2.22	0.02	0.01	0.01	1.36	31.30	0.49	0.43	0.06	0.88	0.12
	W	X	2	1.71E-03	1.71E-03	2.86	0.03	0.01	0.01	1.35	29.10	0.49	0.39	0.10	0.80	0.20
	W	2X	1	1.39E-03	9.93E-03	3.03	0.05	0.03	0.02	1.26	30.00	0.52	0.38	0.14	0.72	0.28
	W	2X	2	3.40E-04	1.13E-02	2.35	0.01	0.01	0.00	1.40	31.60	0.47	0.44	0.03	0.94	0.06
DARLINGTON	D	A	1	3.15E-02	7.14E-02	2.13	0.15	0.14	0.14	1.03	17.10	0.61	0.18	0.44	0.29	0.71
	D	A	2	3.52E-02	9.03E-02	2.11	0.13	0.21	0.18	1.10	17.70	0.58	0.19	0.39	0.33	0.67
	D	D	1	4.20E-02	1.08E-01	1.88	0.13	0.10	0.12	1.16	14.90	0.56	0.17	0.39	0.31	0.69
	D	D	2	4.01E-02	1.00E-01	2.36	0.14	0.11	0.15	1.12	15.50	0.58	0.17	0.40	0.30	0.70
	D	X	1	2.90E-02	6.29E-02	1.92	0.12	0.08	0.10	1.16	18.10	0.56	0.21	0.35	0.37	0.63
	D	X	2	2.38E-02	4.86E-02	1.90	0.17	0.07	0.08	0.85	21.80	0.68	0.19	0.49	0.27	0.73
	D	2X	1	1.48E-02	3.44E-02	1.56	0.15	0.07	0.05	0.88	27.70	0.67	0.24	0.43	0.36	0.64
	D	2X	2	9.49E-03	3.95E-02	1.87	0.08	0.09	0.06	1.23	24.10	0.54	0.29	0.24	0.55	0.45
PICKERING	P	A	1	3.10E-02	7.21E-02	2.23	0.15	0.12	0.13	0.98	20.60	0.63	0.20	0.43	0.32	0.68
	P	A	2	4.18E-02	1.02E-01	2.44	0.14	0.16	0.20	0.86	31.10	0.67	0.27	0.41	0.40	0.60
	P	D	1	4.48E-02	1.45E-01	1.63	0.11	0.14	0.13	1.03	29.90	0.61	0.31	0.31	0.50	0.50
	P	D	2	2.80E-02	7.80E-02	1.77	0.14	0.16	0.12	0.99	23.40	0.63	0.23	0.40	0.37	0.63
	P	X	1	2.25E-02	8.04E-02	1.76	0.10	0.08	0.07	0.90	42.20	0.66	0.38	0.28	0.57	0.43
	P	X	2	1.48E-02	6.95E-02	1.61	0.07	0.08	0.07	1.02	39.20	0.61	0.40	0.21	0.65	0.35
	P	2X	1	1.45E-02	6.90E-02	1.84	0.07	0.03	0.04	0.95	45.50	0.64	0.43	0.21	0.67	0.33
	P	2X	2	7.94E-03	6.99E-02	2.63	0.05	0.06	0.06	0.99	47.40	0.63	0.47	0.16	0.75	0.25
N. BONNARVILLE-ORG	O	A	1	3.20E-03	2.67E-02	3.33	0.04	0.03	0.03	1.19	36.40	0.55	0.43	0.12	0.78	0.22
	O	A	2	3.86E-03	2.41E-02	3.24	0.05	0.04	0.04	1.22	30.50	0.54	0.37	0.16	0.69	0.31
	O	D	1	3.12E-03	2.60E-02	4.22	0.04	0.05	0.05	1.10	42.20	0.58	0.46	0.12	0.79	0.21
	O	D	2	1.60E-03	5.52E-03	8.13	0.10	0.06	0.08	0.89	42.20	0.67	0.37	0.29	0.56	0.44
	O	X	1	1.09E-03	2.18E-02	4.48	0.02	0.02	0.02	1.24	39.20	0.53	0.48	0.05	0.91	0.09
	O	X	2	1.21E-03	2.02E-02	3.10	0.02	0.01	0.01	1.00	56.70	0.62	0.57	0.06	0.91	0.09
	O	2X	1	7.22E-04	3.60E-03	5.43	0.06	0.02	0.02	0.96	46.70	0.64	0.45	0.19	0.71	0.29
	O	2X	2	9.03E-04	8.21E-03	3.92	0.04	0.01	0.01	0.99	52.10	0.63	0.51	0.11	0.82	0.18
N BONNARVILLE-SAND	S	A	1	5.12E-02	2.13E-01	1.51	0.08	0.10	0.11	1.54	11.90	0.42	0.18	0.24	0.44	0.56
	S	A	2	4.13E-02	1.42E-01	1.61	0.10	0.08	0.09	1.46	11.00	0.45	0.16	0.29	0.36	0.64
	S	D	1	6.69E-02	2.31E-01	1.61	0.10	0.12	0.14	1.50	9.62	0.43	0.14	0.29	0.33	0.67
	S	D	2	5.21E-02	2.08E-01	1.38	0.09	0.08	0.09	1.65	7.66	0.38	0.13	0.25	0.33	0.67
	S	X	1	5.68E-03	4.06E-02	2.09	0.05	0.02	0.02	1.61	15.70	0.39	0.25	0.14	0.65	0.35
	S	X	2	2.62E-03	2.17E-02	2.97	0.04	0.02	0.02	1.59	17.20	0.40	0.27	0.13	0.68	0.32
	S	2X	1	2.69E-03	2.99E-02	2.69	0.03	0.02	0.02	1.50	22.60	0.43	0.34	0.09	0.78	0.22
	S	2X	2	5.31E-03	2.79E-02	2.51	0.06	0.03	0.03	1.39	20.30	0.48	0.28	0.19	0.59	0.41

Appendix 3b (cont'd)

Derived HT Oxidation Rate k, Soil Porosities and Fraction of Pore Space with Air and Water

Site	Code	Soil Treatment	Replicate	kN (1/s)	k=kN/Ap (1/s)	Ze*1E2 (m)	Deff*1E4 [Do*AP/T] (m^2/s)	Deff*1E4 [Vg2/kN] (m^2/s)	Deff*1E4 [vg*Ze] (m^2/s)	BD*1E3 (kg/m^3)	%H2O	%Pore Spa	Theta W	Theta A	FPH2O	FPair
BOLTON	B	A	1	1.06E-02	4.82E-02	2.31	0.07	0.05	0.05	0.95	43.90	0.64	0.42	0.22	0.65	0.35
	B	A	2	1.17E-02	5.05E-02	2.72	0.07	0.08	0.08	0.98	44.30	0.63	0.43	0.20	0.68	0.32
	B	D	1	1.99E-02	6.42E-02	2.31	0.11	0.09	0.10	0.89	39.80	0.67	0.35	0.31	0.53	0.47
	B	D	2	1.95E-02	7.00E-02	2.20	0.09	0.08	0.09	0.98	39.60	0.63	0.39	0.25	0.61	0.39
	B	X	1	7.33E-03	6.11E-02	1.89	0.04	0.02	0.02	1.04	47.30	0.61	0.49	0.12	0.81	0.19
	B	X	2	9.47E-03	5.57E-02	2.26	0.06	0.04	0.04	0.89	55.90	0.67	0.50	0.17	0.75	0.25
	B	2X	1	2.59E-04	2.00E-03	5.00	0.03	0.01	0.01	0.98	55.70	0.63	0.54	0.09	0.86	0.14
	B	2X	2	8.53E-03	4.74E-02	2.38	0.06	0.04	0.05	0.89	54.20	0.67	0.48	0.18	0.72	0.28
CLAREMONT	L	A	1	1.93E-02	8.04E-02	1.72	0.08	0.06	0.06	0.91	45.10	0.66	0.41	0.24	0.63	0.37
	L	A	2	2.65E-02	1.06E-01	1.60	0.09	0.06	0.07	0.91	44.40	0.66	0.41	0.25	0.62	0.38
	L	D	1	2.83E-02	8.32E-02	1.81	0.12	0.09	0.09	0.81	43.00	0.69	0.35	0.34	0.50	0.50
	L	D	2	2.62E-02	6.72E-02	2.03	0.13	0.09	0.10	0.81	37.40	0.69	0.30	0.39	0.44	0.56
	L	X	1	7.30E-03	3.65E-02	2.01	0.07	0.04	0.04	0.86	55.50	0.67	0.48	0.20	0.71	0.29
	L	X	2	8.36E-03	3.48E-02	2.35	0.08	0.04	0.04	0.79	58.60	0.70	0.46	0.24	0.66	0.34
	L	2X	1	5.64E-03	3.32E-02	2.27	0.06	0.05	0.04	0.80	65.50	0.70	0.52	0.17	0.75	0.25
	L	2X	2	5.16E-03	3.04E-02	2.17	0.06	0.02	0.02	0.81	64.30	0.69	0.52	0.17	0.75	0.25
GLEN MAJOR	G	A	1	2.67E-02	6.36E-02	1.88	0.14	0.08	0.09	0.63	55.70	0.76	0.35	0.42	0.46	0.54
	G	A	2	2.60E-02	7.22E-02	1.77	0.12	0.08	0.08	0.78	44.30	0.71	0.34	0.36	0.49	0.51
	G	D	1	3.16E-02	8.54E-02	2.12	0.13	0.13	0.14	0.83	38.00	0.69	0.31	0.37	0.46	0.54
	G	D	2	3.11E-02	7.07E-02	2.31	0.15	0.17	0.17	0.78	34.80	0.71	0.27	0.44	0.38	0.62
	G	X	1	5.57E-03	1.99E-02	2.77	0.10	0.05	0.04	0.78	55.80	0.71	0.43	0.28	0.61	0.39
	G	X	2	8.50E-03	2.50E-02	2.63	0.12	0.06	0.06	0.75	50.80	0.72	0.38	0.34	0.53	0.47
	G	2X	1	7.57E-03	3.79E-02	2.40	0.07	0.05	0.04	0.80	62.60	0.70	0.50	0.20	0.72	0.28
	G	2X	2	4.85E-03	4.04E-02	2.63	0.04	0.03	0.03	0.80	72.50	0.70	0.58	0.12	0.83	0.17
BALLANTRAE	A	A	1	6.60E-02	1.53E-01	1.74	0.15	0.16	0.18	1.09	14.80	0.59	0.16	0.43	0.27	0.73
	A	A	2	2.41E-02	5.74E-02	2.75	0.14	0.18	0.18	1.11	14.50	0.58	0.16	0.42	0.28	0.72
	A	D	1	4.04E-02	9.40E-02	1.89	0.15	0.12	0.13	1.07	15.60	0.59	0.17	0.43	0.28	0.72
	A	D	2	3.92E-02	1.03E-01	1.93	0.13	0.13	0.14	1.20	14.20	0.55	0.17	0.38	0.31	0.69
	A	X	1	2.63E-02	7.97E-02	1.83	0.11	0.08	0.08	1.14	21.50	0.57	0.24	0.33	0.43	0.57
	A	X	2	2.06E-02	7.36E-02	2.19	0.10	0.09	0.09	1.15	25.00	0.57	0.29	0.28	0.51	0.49
	A	2X	1	8.28E-03	3.07E-02	2.10	0.09	0.03	0.04	1.06	31.10	0.60	0.33	0.27	0.55	0.45
	A	2X	2	9.08E-03	3.49E-02	2.31	0.09	0.05	0.05	1.11	28.70	0.58	0.32	0.26	0.55	0.45
KINSALE	K	A	1	2.17E-02	7.75E-02	1.95	0.10	0.07	0.08	0.93	40.60	0.65	0.38	0.28	0.58	0.42
	K	A	2	3.28E-02	1.17E-01	2.16	0.10	0.13	0.14	0.91	41.40	0.66	0.38	0.28	0.58	0.42
	K	D	1	1.87E-02	7.19E-02	1.71	0.09	0.05	0.05	1.05	33.00	0.60	0.35	0.26	0.57	0.43
	K	D	2	2.54E-02	8.19E-02	2.05	0.11	0.10	0.10	0.94	36.30	0.65	0.34	0.31	0.53	0.47
	K	X	1	1.20E-02	4.44E-02	2.00	0.09	0.04	0.05	0.86	46.80	0.67	0.40	0.27	0.60	0.40
	K	X	2	8.28E-03	4.36E-02	1.86	0.06	0.03	0.03	0.95	47.70	0.64	0.45	0.19	0.71	0.29
	K	2X	1	1.06E-03	9.79E-03	2.53	0.06	0.01	0.01	0.89	53.50	0.67	0.47	0.19	0.71	0.29
	K	2X	2	3.68E-03	3.07E-02	2.27	0.04	0.02	0.02	1.01	49.40	0.62	0.50	0.12	0.81	0.19

Note: Bulk density (BD), %H2O (% water with respect to soil dry wt.), Theta W and Theta A (fraction of soil volume filled by water and air, respectively), and FPH2O and FPair (fraction of pore volume filled by water and air, respectively) are all for 0-0.12 m (0 -12 cm) core depth.

APPENDIX 4

TRITIUM BALANCE FOR CHAMBER EXPERIMENTS ON TRITIUM DEPOSITION

Date (1985)	Site	$[HT]_{initial}$ (kBq)	$[HT]_{final}$ (kBq)	Exposure Time (min)	ΣHTO in Soil Cores (kBq)	HTO in Rim Water (kBq)	% Difference (I-F)/I X 100*
Aug. 28	Cherrywood Wesleyville	7660	6610	39.5	614 311	18.5	1.4
Aug. 28	Darlington Pickering	7130	6340	25.8	500 522	18.5	3.5
Sept. 4	Bowman.-Org. Bowman.-Sand Bolton	7000	6080	43.5	118 625 370	14.8	3.0
Sept. 11	Claremont Glen Major	7000	5950	49.0	514 648	7.4	1.7
Sept. 13	Ballantrae Kinsale	7130	6340	32.2	640 352	7.4	2.9

*Tritium Balance: $I = [HT]_{initial}$

$F = [HT]_{final} + \Sigma$[HTO in Soil Cores] + [HTO in Rim Water]